

MASS TRANSFER AND SEPARATION OF
SPECIES IN OSCILLATING FLOWS

By

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To Susan, Yesenia, and Dakota.

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LIST OF SYMBOLS

a	Geometric spacing, length. Applies to Equations 1.1 and 1.2.
a	radius of a tube, length. Applies to Equations 2.44 – 2.56.
A	Peak-to-peak amplitude, cm.
A, A^*	The operator and its adjoint. Applies to Equations 2.66 and 2.67.
b	Length scale in semi-infinite geometry, cm.
c	Species concentration, mol/cm ³ .
c_1, c_2	Species concentration in Reservoir 1 and Reservoir 2 respectively, mol/cm ³ .
C_n	Concentration moments, mol/cm ³ . Applies to section 2.4.
D	Molecular diffusion coefficient, cm ² /sec.
h	Plate spacing, cm.
$i = \sqrt{-1}$	Imaginary number.
J	Molar flux, mol/(cm ² *sec).
L	Plate length or tube length, cm.
O, O'	Fixed observer and observer on the moving frame respectively.
P	Pressure, g/(cm*sec ²).
q	Flow rate in two-dimensions, cm ² /sec.

Q	Mass transfer, $\text{mol}/(\text{cm}^2 \cdot \text{sec})$ in two-dimensions and mol/sec in three-dimensions.
r	radial coordinate or radial component in circular geometry.
R	Cross-sectional area, length^2 . Applies to Equation 1.2.
R	Radius for the open tube, cm.
R_{in}	Inner radius for the annular configuration, cm.
R_{out}	Outer radius for the annular configuration, cm.
S	Second order solution to the concentration in eccentric problem.
$Sc = \nu/D$	Schmidt number, dimensionless.
t	time, sec.
u	Velocity, cm/sec . Applies to section 2.4.
U	Second order solution to the velocity in eccentric problem.
V	Velocity, cm/sec .
V	Tidal volume, length^3 . Applies to Equation 1.2.
$W = b(\omega/\nu)^{1/2}$	Womersley number in semi-infinite geometry, dimensionless.
$W = a(\omega/\nu)^{1/2}$	Womersley number, dimensionless. Equations 1.1 and 1.2.
$\mathcal{W} = h(\omega/\nu)^{1/2}$	Womersley number, dimensionless. Applies to sections 2.1 and 2.2.
$W = h(\omega/2\nu)^{1/2}$	Womersley number, dimensionless. Applies to section 2.3.
$\hat{W} = (i + 1)W$	Complex Womersley number, dimensionless.
$\tilde{W} = (i - 1)W$	Complex Womersley number, dimensionless.
$W = R(\omega/\nu)^{1/2}$	Womersley number for an open tube, dimensionless.
$W = R_{\text{out}}(\omega/\nu)^{1/2}$	Womersley number for an annulus, dimensionless.

x	x-coordinate or component in two dimensions.
y	y-coordinate or component in two dimensions.
z	axial coordinate or axial component in circular geometry.

Greek Symbols

$\alpha = (i\omega/D)^{1/2}$	Diffusive length grouping to aid in derivations, cm.
β	Constant in solutions to velocity equations.
ϵ	Eccentric displacement, cm.
ϕ	Phase angle, radians.
η	Constants in solutions to concentration equations.
φ	Constants in solutions to concentration equations.
φ_j	Eigenfunctions. Applies to section 2.4.
$\lambda = (i\omega/\nu)^{1/2}$	Viscous length grouping to aid in derivations, cm.
μ	Dynamic viscosity, g/(cm*sec).
ν	Kinematic viscosity, cm ² /sec.
θ	Angular coordinate or angular component in circular geometry.
ρ	Density, g/cm ³ .
ω	Frequency of oscillation, rad/sec.
ξ	Oscillating piston position, cm.
ψ	Constants in solutions to velocity equations.
ψ_j	Eigenfunctions. Applies to section 2.4.

Subscripts

eff	Effective value.
diff	Diffusive portion.
conv	Convective portion.

Superscripts

* , $\sim$	Complex conjugates.
$\rightarrow$	Denotes a vector.
$\overline{}$	Overbar - time average over one period.
$\overline{}$	Overbar - Spatial average (mean). Applies to section 2.4.
tavg	Time average over one period. Applies to section 2.4.

Abstract of Dissertation Presented to the Graduate School
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SPECIES IN OSCILLATING FLOWS

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Oscillating flows have been shown to greatly enhance the transport of a dilute species over that due to pure molecular diffusion. Although there is no net flow in a periodic cycle, the motion of the fluid creates radial concentration gradients that allow a dilute species to move in a zig-zag fashion from the core of a tube to the boundary and back again while being convected axially along the tube. If two dilute species are present in a carrier, the rate of movement of each species in relation to the fluid motion will be different because of the differing diffusion coefficients. In fact, depending on the diffusion coefficient, the kinematic viscosity of the carrier, and the frequency of oscillation, the faster diffusing species may have a higher, a lower, or the same total mass transfer as the slower diffusing species. As the mass transfer of each species is different, a separation can thus be effected.

This dissertation is a study on the physics behind the phenomena of oscillatory flows in several geometries. Flow in an infinite fluid above an oscillating plate, flow

between two flat plates, flow in an open tube, and flow in the annular region between two tubes have all been examined in terms of overall mass transfer and separation of dilute species. For annular periodic flow, configurations where the tubes oscillate out of phase, tubes oscillating at different frequencies, and an eccentric annulus where the inner tube is slightly off center from the outer tube have also been studied.

Experiments have been performed to verify the accuracy of the models developed in this study. The models closely predicted the overall trends and the physics of the phenomena that occurred in experimental runs, especially the separation of the dilute species obtained experimentally. However, the models are insufficient in some aspects in predicting the complicated flows that are present in the experiments. These findings are presented along with discussions on how the models or the experimental apparatus can be improved to accurately predict the separation and mass transfer of dilute species in oscillating flows.

CHAPTER 1 INTRODUCTION OF THE PHYSICS

1.1 Physics of the Fluid Flow--Mass Transport Interaction

Picture a tube with a gas occupying a reservoir at each end. One of the reservoirs can be considered to hold a pure gas called the carrier while the other is a mixture of the carrier gas with a dilute amount of a single species as shown in Fig. 1.1. According to Fick's law for dilute species, the non-interacting particles will move from the mixture to the carrier gas from high to low concentration in a process involving pure molecular diffusion. Now, suppose that the fluid in the tube oscillates with no net flow from one reservoir to the other. This, for example, can be achieved with a piston. In the first half of a cycle of the piston stroke, a nearly parabolic flow profile is produced in the tube, provided that the frequency is not large. This in turn causes radial concentration gradients. The dilute species then diffuses from the core of the tube to the boundary. In the second half of a cycle, the flow profile is reversed and the species moves from the boundary to the core where the concentration of the species is small compared to the boundary. In the first half of the next cycle of the piston stroke, the species which is in the fast moving core of the tube is convected down the tube and again radially diffuses toward the wall of the tube. The species thus proceeds to move in this zigzag fashion down the tube giving a higher transport of it than by pure molecular diffusion, yet with no net flow between the two reservoirs. If another non-interacting dilute species were

added to the mixture, the time constants of the system become very important due to the differing diffusion coefficients of the species, the frequency of oscillation, and the

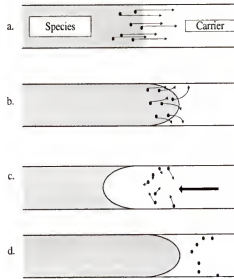


Figure 1.1 Dispersion mechanism for dilute species in pulsatile flow.

kinematic viscosity of the fluid. These different time constants can give rise to a separation of species due to the periodic flow. It is the relationship of the different time constants that govern the mass transfer for each species as well as an analysis of the mass transfer and separation of species for different geometries that are the focus of this research.

1.2 Key Objectives

Periodic flow greatly enhances the mass transfer of a species over that due to pure molecular diffusion. The primary goal is to understand this enhancement and the separation that can be achieved through the key operating and thermophysical parameters that arise in periodic flow problems. Because of the complexity of the system, simplified

geometries such as an oscillating plate in an infinite fluid and periodic flow in a channel will help in the theoretical understanding of the physics of the problem. A simplified geometry would make the calculations easier and the subsequent results easier to interpret than those in a more complicated geometry. The overall trends and phenomena observed in flow between flat plates will not change in examining periodic flow in a circular geometry; only quantitative results would be different.

Experiments will also be used to verify the phenomena observed theoretically. Because of the experimental setup, periodic flow in a tube and in an annular region between two tubes will be examined. A detailed explanation of the physics will not be given for these geometries because they are similar to flow in a flat plate; however, theoretical results calculated in this dissertation and by those preceding this research [1] will be verified.

In order to help explain discrepancies that may arise between theoretical predictions and experimental results, certain calculations related to the geometry of the annular system will be performed. Specifically, an eccentricity calculation will be done to see how the mass transfer and separation are affected if the inner tube is slightly off center. Also, tubes oscillating out of phase or at different amplitudes will be examined. Although these configurations may appear to be imperfections in the system, they may actually help in the mass transfer, the separation, or both.

1.3 Historical Perspective

Dispersion in periodic flow has its roots in Taylor's paper [2] demonstrating dispersion in steady flow. Taylor proved that even though convection is important in the

dispersion of a solute, molecular diffusion, especially radial diffusion towards the boundary, could not be totally neglected to describe phenomena observed experimentally. For steady flow, Taylor observed that the effective diffusion coefficient of a system is proportional to the square of the maximum velocity and spacing and *inversely proportional* to the molecular diffusion coefficient. In other words, the longitudinal dispersion was shown to be inversely proportional to the diffusivity. This idea would later prove to be central to the understanding of separation between two species, one with a small diffusion coefficient and the other with a large diffusion coefficient.

Many have used Taylor's discovery as a basis in modeling the mass transport of a solute in periodic flow. Harris and Goren [3] were among the first researchers to prove that periodic flow driven by an oscillating piston can greatly enhance the mass transport of a dilute solute over pure molecular diffusion. This mass transport increases for increasing frequency which in scaled form is given by the Womersley number. This number is defined as

$$W = a\sqrt{\frac{\omega}{\nu}} \quad (1.1)$$

where a is the spacing, ν is the kinematic viscosity, and ω is the oscillation frequency. Although there is no net flow as in the steady flow case, a net transport of a dilute species can still be achieved as previously explained. Harris and Goren obtained an expression for the convective mass transfer scaled with respect to the transport due to molecular diffusion alone. They showed that this was proportional to the square of the ratio of the stroke amplitude to plate spacing and a function of the Womersley number and Schmidt number in a complicated way. These findings were backed by experiments in liquids using HCl in an aqueous solution.

Watson [4] derived like expressions for periodic flow in general geometries and in particular presented calculations for the special cases of flow in a circular pipe and in a two-dimensional channel. He was able to show that the effective diffusion coefficient was of the form

$$\frac{D_{eff}}{D} = 1 + f(W, Sc) \left(\frac{V}{aR} \right)^2 \quad (1.2)$$

where Sc is the Schmidt number, R is the cross-sectional area, V is the tidal volume for the oscillations, and D is the molecular diffusion coefficient. Watson's results were verified experimentally in a companion paper by Joshi et al. [5].

Kurzweg, Jaeger, and fellow collaborators [6-11] have performed extensive research in the area of periodic flow. Most significantly, they observed that for a fixed frequency, the system can be "tuned" to a specific Womersley number to achieve the greatest effective diffusion coefficient [7]. This tuning occurs when the molecular diffusion length for a given radius of the tube is on the same order as one half of the period or

$$\frac{a^2}{D} = \frac{\pi}{\omega} \quad (1.3)$$

which can be written in dimensionless terms as

$$W^2 Sc = \pi \quad (1.4)$$

This implies that if the tube radius is very small for a given oscillation frequency, most of the dilute species is able to diffuse radially to the wall before the reverse piston stroke. Being near the wall of the tube in the viscous boundary layer means more of the species is in a slow moving region of the flow and does not fully take advantage of the higher velocity in the core of the tube. On the other hand, if the tube radius is large for a given

frequency, most of the fluid exhibits a plug flow profile except for a small region where the velocity drops sharply near the wall. If most of the tube exhibits plug flow, radial concentration gradients are not utilized in these regions making it inefficient. "Tuning" the process requires operation in between these two extremes.

It can be seen by the above relation that tuning takes place at different Womersley numbers for different species because of the difference in Schmidt number. Tuning for liquid species and gaseous species will naturally be different. Jaeger et al. [10] have indeed shown this to be the case for a system of CO_2 dispersion in an O_2 carrier with a Schmidt number equal to 1 and He in O_2 system with a Schmidt number equal to 0.2. The tuning occurs at a smaller Womersley number for the CO_2 system and yields a higher effective diffusion coefficient. The difference in the tuning and the value of the effective diffusion coefficient can be used to achieve an optimal region of separation of the two species. For a liquid system of $Sc = 2000$ and $Sc = 500$, they showed that the lighter species ($Sc = 2000$) had a maximum effective diffusion coefficient at a smaller Womersley number and was of the same order as the effective diffusion coefficient for the heavier species. They then concluded that for a gas system the effective diffusion coefficient of the heavier species ($Sc = 1$) is higher than that of the light species for all Womersley numbers, whereas for liquids, the heavier species may either have a higher or lower value for the effective diffusion coefficient than the heavier species.

Jaeger et al. [12] also studied the effect of counterflow on the mass transfer and separation of various compositions of O_2 in a helium carrier. Counterflow is defined as a steady flow against the direction of the concentration gradient superimposed upon the oscillatory flow. It was found that the separation achieved with counterflow was 20 to 40

times better than with only oscillating flow. However, it can be surmised that counterflow will decrease a species' overall mass transfer because it flows against the concentration gradient. Optimization is needed for this configuration to appreciably enhance the separation without too much of a decrease in the mass transfer.

A final area of note is the use of pulsatile flow in heat transfer as has been analyzed by Kurzweg and Chen [13]. The governing equations for heat transfer and mass transfer are similar in nature, so one would expect to see similar results in a system involving the removal of heat to a system involving diffusing species.

Further references to past work will be made throughout this dissertation where appropriate.

1.3 A Fluid in an Infinitely Deep Channel Driven by an Oscillating Boundary

A simple calculation of a fluid in a channel, semi-infinite in width and infinitely deep, driven by an oscillating boundary will be used as an introduction to a more complicated system. The velocity and concentration profile for this case reveals some important aspects of periodic flow problems and their effect on the mass transfer and separation of species.

Fig. 1.2 is the depiction of a system where the lower boundary moves periodically in the 'x' direction, and the fluid is of infinite extent in the 'y' direction. The velocity at the wall is assumed to be

$$V_x = \frac{1}{2} A \omega \cos(\omega t) = \frac{1}{4} A \omega [\exp\{i\omega t\} + \exp\{-i\omega t\}] \quad (1.5)$$

where A is the peak-to-peak amplitude and ω is the imposed frequency of motion of the

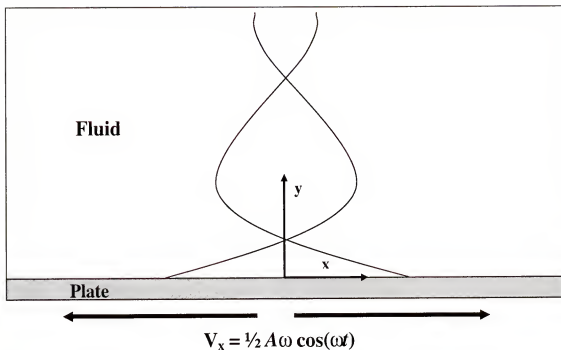


Figure 1.2 Semi – infinite geometry with oscillating plate.

wall. As the far field velocity is zero, we also have

$$V_x = 0 \text{ as } y \rightarrow \infty \quad (1.6)$$

Assuming the fluid to be incompressible, Newtonian, with a kinematic viscosity, ν , and the flow to be laminar, the x -component of the equations of motion for this system is

$$\frac{\partial V_x}{\partial t} = \nu \frac{\partial^2 V_x}{\partial y^2} \quad (1.7)$$

Observe here that a pressure gradient neither drives nor is induced by the flow. Now as the boundary motion of this system is periodic, it is assumed that the velocity is also periodic and of the form

$$V_x = \hat{V}_x e^{i\omega t} + \tilde{V}_x e^{-i\omega t} \quad (1.8)$$

where “ $\hat{}$ ” and “ $\sim$ ” denote complex conjugates. Substituting this form of the velocity into the equation of motion yields

$$\frac{d^2 \hat{V}_x}{dy^2} - \lambda^2 \hat{V}_x = 0 \quad (1.9)$$

and

$$\frac{d^2 \tilde{V}_x}{dy^2} + \lambda^2 \tilde{V}_x = 0 \quad (1.10)$$

where

$$\lambda \equiv \sqrt{\frac{i\omega}{\nu}} \quad (1.11)$$

Solving these equations subject to the above boundary conditions gives

$$\hat{V}_x = \frac{1}{4} A \omega e^{-\lambda y} \quad (1.12)$$

and

$$\tilde{V}_x = \frac{1}{4} A \omega e^{\lambda y} \quad (1.13)$$

Because $V_x = 2\Re(\hat{V}_x e^{i\omega t})$ where $\Re(g)$ means the real part of 'g', we see that

$$V_x = \frac{1}{2} A \omega e^{-y\sqrt{\omega/2\nu}} \cos\left(y\sqrt{\omega/2\nu} - \omega t\right) \quad (1.14)$$

This result can also be found in Schlichting [14], and it is interesting for several reasons.

First, it demonstrates that the velocity profile decays with increasing distance from the oscillating wall and that it decays periodically in space and time. Second, there is a phase lag between the fluid velocity and the wall velocity. This is given by $y\sqrt{\omega/2\nu}$. This

phase lag is a function of ω and ν , and as the frequency increases, the phase lag becomes more apparent provided the kinematic viscosity is small. The distinguished region where

the phase lag is unimportant is where $y \ll \sqrt{2\nu/\omega}$. This phase lag will become an

important issue later in this dissertation as an explanation is given for the difference in the relative mass transfer of the two species.

Now that the velocity of the system has been obtained, the concentration field $c(x,y,t)$ can be calculated by assuming that each species is in dilute concentration with a constant molecular diffusion coefficient, D , that no chemical reactions are present, and that the diffusion flux obeys Fick's law. The modeling equation for species conservation is then

$$\frac{\partial c}{\partial t} + V_x \frac{\partial c}{\partial x} = D \frac{\partial^2 c}{\partial x^2} + D \frac{\partial^2 c}{\partial y^2} \quad (1.15)$$

The boundary conditions that imply that the plate is impermeable to the transport of the dilute species and that there are no concentration gradients in the y -direction far from the moving plate are

$$\frac{\partial c}{\partial y} = 0 \text{ at } y = 0 \quad (1.16)$$

and

$$\frac{\partial c}{\partial y} = 0 \text{ as } y \rightarrow \infty \quad (1.17)$$

Now, to aid in getting a solution, the concentration field in the fluid is assumed to be

$$c = \frac{(c_2 - c_1)x}{L} + \hat{c}(y)e^{i\omega t} + \tilde{c}(y)e^{-i\omega t} \quad (1.18)$$

where $(c_2 - c_1) / L$ is the mean axial concentration gradient.¹ Substituting this form of the concentration into the species conservation equation and separating like exponentials will give one equation with $\hat{c}$ and another with $\tilde{c}$. The solution subject to the boundary

¹ The validity of this approximation will be discussed further in Chapter 2.

conditions is then seen to be

$$\hat{c} = \frac{\frac{1}{4} A \omega (c_2 - c_1)}{DL(\lambda^2 - \alpha^2)} \left(e^{-\lambda y} - \frac{\lambda}{\alpha} e^{-\alpha y} \right) \quad (1.19)$$

and

$$\tilde{c} = \frac{-\frac{1}{4} A \omega (c_2 - c_1)}{DL(\lambda^2 - \alpha^2)} \left(e^{i\lambda y} + \frac{\lambda}{\alpha} e^{i\alpha y} \right) \quad (1.20)$$

where

$$\alpha \equiv \sqrt{\frac{i\omega}{D}} \quad (1.21)$$

The time-averaged flux of the system is found by taking the time average of the instantaneous flux over one time period. It is expressed as the sum of two terms: one corresponding to the usual molecular diffusion in the x -direction and the other due to the transport by convection. Calling the time-averaged flux over one period of oscillation, $\bar{J}$, we see that

$$\bar{J} = -D \frac{\partial \bar{c}}{\partial x} + \overline{V_x c} \quad (1.22)$$

where

$$\overline{V_x c} = \frac{\omega}{2\pi} \int_0^{2\pi} V_x c dt = \tilde{V}_x \tilde{c} + \tilde{V}_x \hat{c} = 2\Re(\tilde{V}_x \hat{c}) \quad (1.23)$$

and consequently

$$\bar{J} = -\frac{D(c_2 - c_1)}{L} + 2\Re(\tilde{V}_x \hat{c}) \quad (1.24)$$

To find the overall time- and space-averaged mass transfer of the species, the time-averaged flux must be integrated across the entire region of interest. Now as the system is a fluid of infinite extent, a length scale b is introduced as the region over which

the majority of the mass transfer is taking place, but its value can only be determined later. The time- and space-averaged overall mass transfer, $\bar{Q}$, is then

$$\bar{Q} = \frac{1}{b} \int_0^b \bar{J} dy = \frac{-D(c_2 - c_1)}{L} + \frac{2}{b} \Re \int_0^b \tilde{V}_x \hat{c} dy \quad (1.25)$$

i.e.,

$$\bar{Q} = \bar{Q}_{diff} + \bar{Q}_{conv} \quad (1.26)$$

where $\bar{Q}_{diff}$ and $\bar{Q}_{conv}$ are easily identified from the above equation as the part due to diffusion and convection respectively. We see in the above formula for $\bar{Q}$ that if b were taken to be infinity then $\bar{Q}_{conv}$ would be zero. On the other hand, if the overall mass transport were not spatially averaged, then the diffusive part of the flux would alone contribute over an infinite extent. If the relative importance of the diffusive and convective flux is to be understood, then the length scale b must be introduced.

Now, returning to the evaluation of $\bar{Q}$ we see that

$$\bar{Q} = \frac{-D(c_2 - c_1)}{L} + \frac{\frac{1}{8} A^2 \omega^2 (c_2 - c_1)}{DLb} \Re \left[\left(\frac{1}{\lambda^2 - \alpha^2} \right) \left(\frac{e^{(i-1)\lambda b} - 1}{(i-1)\lambda} - \frac{\lambda}{\alpha} \frac{e^{(i\lambda - \alpha)b} - 1}{i\lambda - \alpha} \right) \right] \quad (1.27)$$

Substituting the definitions of α and λ and observing that $\sqrt{i} = (i+1)/\sqrt{2}$, the overall mass transfer now becomes

$$\bar{Q} = \frac{-D(c_2 - c_1)}{L} + \frac{\frac{1}{8} A^2 \omega^2 (c_2 - c_1)}{L} \frac{1}{\frac{D}{v} - 1} \Re \left[i \left(\frac{e^{-b\sqrt{\frac{2\omega}{v}}} - 1}{-b\sqrt{\frac{2\omega}{v}}} - \sqrt{\frac{D}{v}} \frac{e^{b\sqrt{\frac{\omega}{2v}} \left(i \left(1 - \sqrt{\frac{v}{D}} \right) \left(\sqrt{\frac{v}{D}} + 1 \right) \right)} - 1}{b\sqrt{\frac{\omega}{2v}} \left[i \left(1 - \sqrt{\frac{v}{D}} \right) - \left(\sqrt{\frac{v}{D}} + 1 \right) \right]} \right) \right] \quad (1.28)$$

Two dimensionless groups arise naturally in this formula. One group is the ratio of viscous to diffusive effects or the Schmidt number, $Sc = \nu/D$, and the other is a

dimensionless frequency, or a form of the Womersley number, $W = b(\omega/\nu)^{1/2}$. One also observes that the convective portion has a quadratic dependence on the amplitude, A .

Turning now to the value of b , one may choose it such that the convective mass transfer will be at least 99.9% of the mass transfer if b were only taken to be ∞ . From Equation 1.25, if b were infinite, then the convective mass transfer would be zero. For widths greater than the chosen value of b , the mass transfer is small enough that it will only account for 0.1% of the total mass transfer and can thus be neglected. With this definition of b , we can take the asymptotic limit of $\bar{Q}$ for large b to see that if $\nu < D$ then

$$b \geq \frac{7}{2} \sqrt{\frac{2D}{\omega}} \quad (1.29)$$

and if $\nu > D$, as in the case of liquids, then

$$b \geq \frac{7}{2} \sqrt{\frac{2\nu}{\omega}} \quad (1.30)$$

to ensure that at least 99.9% of the mass transfer is calculated.

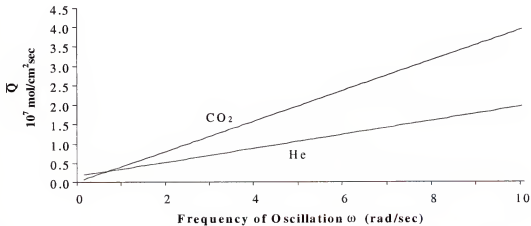


Figure 1.3 Total mass transfer versus frequency for carbon dioxide and helium in a nitrogen carrier in an oscillating flat plate configuration.

When two dilute species are present in the mixture, the total mass transfer is calculated separately for each species assuming that no interactions between the dilute species take place. As an example, Fig. 1.3 is a plot of $\bar{Q}$ which shows the total time- and space-averaged mass transfer versus the frequency for the case of a dilute mixture of helium and CO₂ in a nitrogen carrier gas. Notice that, for high frequencies, the total mass transfer of the slower diffusing species (CO₂) actually has a higher mass transfer than the faster diffusing helium. We can explain this in two ways, one from rewriting the formula for $\bar{Q}$ and the other from a physical perspective.

The above formula for $\bar{Q}$ may be rewritten for the case when $\nu < D$ and

$$b = \frac{1}{2} \sqrt{2D/\omega} \text{ as}$$

$$\bar{Q} = \frac{-D(c_2 - c_1)}{L} - \frac{\frac{1}{56} A^2 \omega (c_2 - c_1)}{\left(\frac{1}{Sc} - 1\right)(1 + Sc)L} \left\{ e^{\frac{7}{2} \left(\frac{1}{\sqrt{Sc}} + 1\right)} \left[\begin{aligned} &\left(1 + \sqrt{Sc}\right) \sin \left[\frac{7}{2} \left(\frac{1}{\sqrt{Sc}} - 1 \right) \right] \\ &+ \left(1 - \sqrt{Sc}\right) \cos \left[\frac{7}{2} \left(\frac{1}{\sqrt{Sc}} - 1 \right) \right] \end{aligned} \right] \right\} + 1 - \sqrt{Sc} \quad (1.31)$$

From this formula for $\bar{Q}$ we see immediately that $\bar{Q}$ is linear in ω with an intercept of

$$\frac{D(c_1 - c_2)}{L}. \text{ The ratio of the slopes of } \bar{Q} \text{ for the two species is approximately of the}$$

order

$$\frac{1 - \sqrt{Sc_1}}{\left(\frac{1}{\sqrt{Sc_1}} - 1\right)(1 + Sc_1)} \frac{\left(\frac{1}{\sqrt{Sc_2}} - 1\right)(1 + Sc_2)}{1 - \sqrt{Sc_2}} \quad (1.32)$$

where Sc_1 and Sc_2 are the Schmidt numbers for the two species. If $D_1 > D_2$, making $Sc_1 < Sc_2$, and Schmidt numbers are less than one as they are in the cases studied here, the ratio

of the slopes is less than unity indicating the slope for the “slow diffuser” is greater than the slope of the “fast diffuser.” Because the values of the intercepts are proportional to the diffusion coefficients, we see at once that both graphs must intersect, and the slow diffuser, i.e. CO_2 , will ultimately transport faster than the fast diffuser (He). But there is a physical explanation for all of this that is now put forth.

As the plate moves, there is a fast flow region near it, and the flow regime becomes slow further away from the boundary. Because of the radial concentration gradients produced by the flow, the faster diffusing species moves more into the slower flow region while the slower diffusing species remains closer to the plate. This allows the slower diffuser to take advantage of the forced convection and thus have the higher total mass transfer.

Of course, the value of b is not the same for both species as it depends on the diffusion coefficient, at least for gases, and as the frequency grows larger, b becomes smaller. This turns out to be important as the mass transfer is averaged over the region b . Although this calculation gives an idea as to how this type of arrangement works, it prompts the question about how the periodic flow with different species affects the mass transport in a system of finite depth where the notion of different b 's does not arise and where the mass transfer must be calculated over the same fixed region. Indeed, we might conjecture that $\overline{Q}$ will not vary linearly with ω in a finite width geometry, and it is to the case of the finite fluid depth in both two-dimensional and circular geometries that is the focus of this work.

CHAPTER 2 PERIODIC FLOW IN A CHANNEL

The first finite geometry that is studied is the periodic flow in a channel between two flat plates. Although this is still a simplified geometry, the physics behind periodic flow and its effect on the mass transfer and separation of species are seen without much of the complicated mathematics associated with a more complicated geometry. The mass transfer and separation will be analyzed along with an explanation of the physics associated with the phenomena observed. Further, periodic flow can be produced using an oscillating pressure drop or by an oscillating boundary. The oscillating pressure drop will be introduced because the explanations regarding the physics are relatively easier to understand. The relationship between a pressure driven and boundary driven configuration will be presented later in the chapter.

2.1 Periodic Flow Between Two Flat Plates a Finite Distance Apart

Fig. 2.1 shows the set up of a fluid driven by pistons between two flat plates ensuring that no net flow is produced over a cycle along the tube for all conditions and parameters of the system. As the figure suggests, the physics of the problem is two-dimensional. For the pressure driven case, the fluid flow is induced by pistons that oscillate according to $\xi = \frac{1}{2} A \sin(\omega t)$ where ξ is the position of both pistons that are in phase. The flow rate q of this system is then

$$q = h \frac{d\xi}{dt} = \frac{1}{2} A \omega h \cos(\omega t) = \frac{1}{4} A \omega h [\exp\{i\omega t\} + \exp\{-i\omega t\}] \quad (2.1)$$

where h is the distance from the top to the bottom plate. Assuming an incompressible, Newtonian fluid, the corresponding equation of motion in the 'x' direction is then

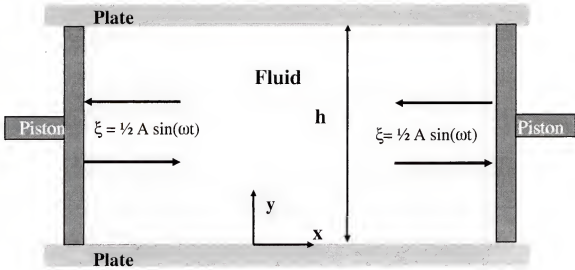


Figure 2.1 Periodic flow between two flat plates produced by two pistons oscillating in phase.

$$\frac{\partial V_x}{\partial t} = -\frac{\Delta P}{L} + \nu \frac{\partial^2 V_x}{\partial y^2} \quad (2.2)$$

provided that the entrance and end effects are neglected. It may be assumed that the velocity of the system is periodic and that the pressure is of the form

$$\frac{\Delta P}{L} = \hat{P}e^{i\omega t} + \tilde{P}e^{-i\omega t} \quad (2.3)$$

With the no-slip boundary condition at the walls of the plate, the expression for the velocity that satisfies the given flow rate is

$$V_x = 2\Re(\tilde{V}_x e^{-i\omega t}) \quad (2.4)$$

with

$$\tilde{V}_x = \frac{1}{4} A \omega \left(\psi_1 e^{i\sqrt{\frac{\omega}{\nu}} y} + \psi_2 e^{-i\sqrt{\frac{\omega}{\nu}} y} + \psi_3 \right) \quad (2.5)$$

ψ_n 's are independent of y and depend only on the thermophysical properties and parameters of the system, ν , ω , and h , and are given in Appendix A.

The concentration field can be found by solving the species continuity equation,

that is

$$\frac{\partial c}{\partial t} + \frac{\partial c}{\partial x} V_x = D \frac{\partial^2 c}{\partial x^2} + D \frac{\partial^2 c}{\partial y^2} \quad (2.6)$$

where Fick's law is assumed to hold. The boundary conditions that imply that the plate is impermeable to the transport of the dilute species are

$$\frac{\partial c}{\partial y} = 0 \text{ at } y = 0 \text{ and } y = h \quad (2.7)$$

Now, to enable us to get a solution, the concentration field in the fluid is assumed to be the sum of two parts, one due to a mean axial concentration gradient and the other due to a transverse variation that also fluctuates with time. This implicitly assumes that the end effects are neglected and that the calculation can be considered to be the same as examining the middle portion of the tube thereby giving us a heuristic model.¹ The concentration field is then written as

$$c = \frac{(c_2 - c_1)x}{L} + c^*(y, t) \quad (2.8)$$

where $(c_2 - c_1)/L$ is the mean axial concentration gradient. The solution to Equation 2.6 subject to the boundary conditions is then

$$c = \frac{(c_2 - c_1)x}{L} + 2\Re\{\hat{c}(y)e^{i\omega t}\} \quad (2.9)$$

with

$$\hat{c} = \frac{1}{4} A \frac{(c_2 - c_1)}{L} \left(\varphi_1 e^{-\sqrt{\frac{\omega}{D}} y} + \varphi_2 e^{\sqrt{\frac{\omega}{D}} y} + \varphi_3 e^{-\sqrt{\frac{\omega}{D}} y} + \varphi_4 e^{\sqrt{\frac{\omega}{D}} y} + \varphi_5 \right) \quad (2.10)$$

¹ This assumption can be justified by the method of moments and the model is similar to the one developed by Harris and Goren [3], Watson [4], and Kurzweg and Jaeger [8] among others. This also assumes that the amplitude to height ratio is large.

where ϕ_n 's are also independent of y and are given in Appendix A.

The time- and space-averaged flux of the system is found by integrating the time averaged flux over the entire distance between the two plates; i.e.,

$$\bar{Q} = -\frac{D(c_2 - c_1)}{L} + \frac{2}{h} \Re \left[\int_0^h \tilde{V}_x \hat{c} dy \right] \quad (2.11)$$

The final expression for the total mass transfer is then

$$\bar{Q} = -\frac{D(c_2 - c_1)}{L} - \frac{\frac{1}{8} A^2 \omega (c_2 - c_1)}{L h \sqrt{\frac{\omega}{\nu}}} \Re \left[i^{3/2} \left(\frac{\psi_1 \phi_1 \left(e^{i \left(\sqrt{\frac{\omega}{\nu}} - \sqrt{\frac{\omega}{D}} \right) h} - 1 \right) - \psi_2 \phi_2 \left(e^{\left(\sqrt{\frac{\omega}{D}} - i \sqrt{\frac{\omega}{\nu}} \right) h} - 1 \right)}{i - \sqrt{\frac{\nu}{D}}} \right. \right. \\ + \frac{\psi_1 \phi_2 \left(e^{i \left(\sqrt{\frac{\omega}{\nu}} + \sqrt{\frac{\omega}{D}} \right) h} - 1 \right) - \psi_2 \phi_1 \left(e^{-i \left(\sqrt{\frac{\omega}{D}} + \sqrt{\frac{\omega}{\nu}} \right) h} - 1 \right)}{i + \sqrt{\frac{\nu}{D}}} \\ + \frac{\psi_1 \phi_3 \left(e^{(i-1) \sqrt{\frac{\omega}{\nu}} h} - 1 \right) - \psi_2 \phi_4 \left(e^{(1-i) \sqrt{\frac{\omega}{\nu}} h} - 1 \right)}{i - 1} \\ + \frac{\psi_1 \phi_4 \left(e^{(i+1) \sqrt{\frac{\omega}{\nu}} h} - 1 \right) - \psi_2 \phi_3 \left(e^{-(i+1) \sqrt{\frac{\omega}{\nu}} h} - 1 \right)}{i + 1} \\ - i \psi_1 \phi_5 \left(e^{i \sqrt{\frac{\omega}{\nu}} h} - 1 \right) + i \psi_2 \phi_5 \left(e^{-i \sqrt{\frac{\omega}{\nu}} h} - 1 \right) \\ + \sqrt{\frac{D}{\nu}} \psi_3 \phi_2 \left(e^{\sqrt{\frac{\omega}{D}} h} - 1 \right) - \sqrt{\frac{D}{\nu}} \psi_3 \phi_1 \left(e^{-\sqrt{\frac{\omega}{D}} h} - 1 \right) \\ \left. + \psi_3 \phi_4 \left(e^{\sqrt{\frac{\omega}{\nu}} h} - 1 \right) - \psi_3 \phi_3 \left(e^{-\sqrt{\frac{\omega}{\nu}} h} - 1 \right) + \psi_3 \phi_5 h \sqrt{\frac{\omega}{\nu}} \right] \quad (2.12)$$

It is apparent that three dimensionless groups can be identified. These are $h(\omega/\nu)^{1/2}$, also called the Womersley number (W), the Schmidt number, Sc , and the ratio A/h . As the frequency increases, only the convective part becomes dominant, and it varies

quadratically with A while both diffusive and convective terms are directly proportional to the concentration difference.

Beyond this, the above formula gives a complicated relationship between the total mass transfer, the gap width, and the frequency of motion. However, graphs of the mass transfer will assist in delineating the physics, and so we present the results for 3 special systems: He-CO₂, He-CH₄, and He-X in a pure nitrogen carrier gas where X is an unnamed species with a specific diffusivity in the carrier. The first two are systems for which diffusivity and kinematic viscosity data are readily available from the literature and are given in Table 2.1. Although species X may not be a readily identifiable species, as we will presently see, it is being used to help illuminate some of the phenomena that can occur in periodic flow. These systems have been chosen on account of the disparity in the diffusion coefficients of each species with helium being the fastest diffuser, the arbitrary species X being the next fastest diffuser, CH₄ being slower, and CO₂ being the slowest diffusing species of the four. A typical graph needs to be drawn only between $\overline{Q}$ and ω for a fixed set of values of A , h , ν , and D . Such a graph will not qualitatively change from one where $\overline{Q}$ is represented in terms of $\mathcal{W}$ for fixed Sc and A/h . It is, in fact, easier to explain the physics in terms of unscaled, rather than scaled, quantities, and this is now discussed in the next section.

Table 2.1 Values of system and fluid properties used in example calculations

Symbol	Name	Value	Units
h	Distance between plates	2	cm
A	Peak-to-peak amplitude	20	cm
(c1-c2)/L	Concentration difference divided by the plate length	2.23E-08	mol/cm ⁴
ν_{N_2}	Kinematic viscosity of N ₂	0.15	cm ² /sec
ν_{H_2O}	Kinematic viscosity of Water	0.01	cm ² /sec
D_{He}	Diffusion coefficient of He in N ₂	0.710	cm ² /sec
D_X	Diffusion coefficient of X in N ₂	0.500	cm ² /sec
D_{CH_4}	Diffusion coefficient of CH ₄ in N ₂	0.215	cm ² /sec
D_{CO_2}	Diffusion coefficient of CO ₂ in N ₂	0.165	cm ² /sec
$D_{ethanol}$	Diffusion coefficient of Ethanol in Water	1.0E-04	cm ² /sec
$D_{glucose}$	Diffusion coefficient of Glucose in Water	0.5E-04	cm ² /sec

2.2 Results of Some Calculations for $\bar{Q}$ and Their Explanation:

The results of calculating the formula for $\bar{Q}$ versus ω are shown in Fig. 2.2 assuming an amplitude of 20 cm and a width of 2 cm. Observe first that the total mass transfer for both species continually increases for increasing ω . There is nothing to limit the increase in $\bar{Q}$ with increasing ω except for the onset of secondary flows that are disallowed on account of the assumption that the velocity is unidirectional and only a function of 'y' and 't'. The increase in mass transfer with ω may be traced to the flow rate q which is $\frac{1}{2}A\omega h \cos(\omega t)$. A more striking observation and one already seen by earlier workers² is that "crossovers" occur in the total mass transfer in these binary systems. These crossovers are frequencies at which the total mass transfer for each species is the same and therefore reveal that there are frequencies at which the total mass transfer of two different species varies in relation to one another. For example, depending upon the magnitude of the frequency for the 3 systems, a fast diffusing species

² Kurzweg and Jaeger [8] and Jaeger et al. [10] have shown the existence of a crossover frequency where the axial transport is the same. In their 1987 paper [8], there appears to be an existence of a second crossover in their Figure 2 for large Schmidt numbers and

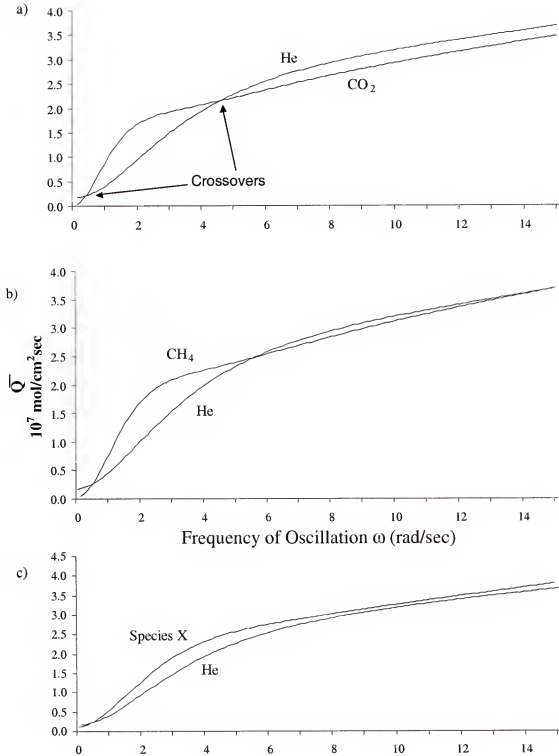


Figure 2.2 Total mass transfer versus frequency for three binary system in a nitrogen carrier. a) He – CO_2 system b) He – CH_4 system c) He – X system

small Womersley numbers. This chapter will demonstrate that there can be up to three crossovers that may occur.

like Helium can either have a lower or higher total mass transfer than the slower diffusing species. The observations are summarized as follows:

- For the He- CO_2 , there exist two crossovers. A middle region occurs where CO_2 has a higher mass transfer than Helium or CH_4 .
- For the He- CH_4 system, three crossovers occur. Helium has a higher mass transfer for small ω , then as ω increases, the mass transfer of CH_4 becomes higher. For yet higher frequencies, Helium regains its earlier position with its mass transfer greater than that for the CH_4 . And finally for very large frequencies, CH_4 has the higher mass transfer.
- For the He-X system, there is only one crossover. Helium initially has the higher mass transfer, but species X overtakes Helium and continues to have the higher mass transfer as frequency increases.

This information can be compressed onto one graph depicting the variations of the ratio

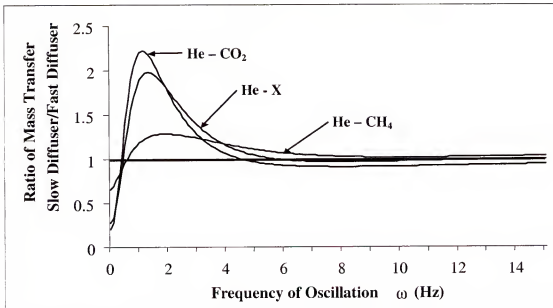


Figure 2.3 Ratio of the total mass transfer of the slow diffuser to the total mass transfer of the fast diffuser for the three binary systems.

of the mass transfer of the slower diffusing species to the faster diffusing species versus frequency. This ratio is plotted for each binary system in Fig. 2.3 with unity indicating the points where the crossovers occur. The mass transfer ratio initially starts at a minimum value at $\omega = 0$ where the fluid is not moving. It then quickly increases to a maximum as the pistons move, decreasing again as ω increases until it reaches a local minimum, and finally slowly increasing for even larger ω 's. All of this demands an explanation, but before one is offered, a digression is made to explain the effect of the diffusion coefficient (D), kinematic viscosity (ν), and frequency of oscillation (ω) on the mass transfer of the system. In fact, these three parameters lead to separate time constants.

The diffusion coefficient, D , can be considered to be a measure of the mobility of a dilute species in a solvent and controls the response time of the transport of a species to a change in concentration. The higher the diffusion coefficient, the faster a species will move in the direction of its concentration gradient, and in periodic flow, the axial and radial mobility is greater for the species with the higher diffusion coefficient.

The kinematic viscosity, ν , controls the response time of the fluid to mechanical disturbances such as the oscillating motion of the piston. The lower the kinematic viscosity, the slower the response time. In periodic flow, low ν can produce "wiggles" in the velocity profile at higher frequencies as the profile noticeably deviates from being parabolic. This means that regions of the fluid flow can be in the forward or positive direction while other regions can have a reverse flow in the negative direction at some given time during one periodic cycle. Higher ν help the fluid maintain its nearly parabolic shape for higher frequencies and gives sharper velocity profiles for lower

frequencies. It is also true that higher ν gives a greater maximum velocity at any given time during a cycle.

The periodicity, $2\pi/\omega$, of the pistons is the amount of time it takes to complete one cycle in periodic flow. Small ω gives the fluid enough time to respond to the oscillating piston, and the species enough time to respond to the changing velocity profile. Although the maximum velocity increases for increasing ω , the amount of time the fluid and each species have to respond to the periodic conditions decreases as each periodic cycle is completed more rapidly.

Returning to the explanation of the shape of the curves in the figures, we see that the primary reason the mass transfer is greater during periodic flow than in a stagnant fluid is the species' ability to take advantage of the forced convection of the fluid to drive it down the tube. The more a species is able to remain near the faster moving region of the fluid, the greater its ability to move down the tube causing greater mass transfer.

Initially, before the onset of the piston movement or when the frequency is zero, the mass transfer of each species is due only to pure molecular diffusion. If the concentration difference between the ends is the same for both species, the faster diffuser will have the higher mass transfer simply because it has the higher diffusion coefficient. The ratio of the mass transfer of the slow diffuser to the fast diffuser is a minimum at zero frequency. Once the piston moves the situation changes. To see why, observe that for small frequencies, a nearly parabolic velocity profile slowly oscillates back and forth with the fastest moving region in the center and the slowest region at the boundary. This profile produces radial concentration gradients that drive both species from the core to the boundary; however, the faster diffuser moves to the slow moving boundary layer

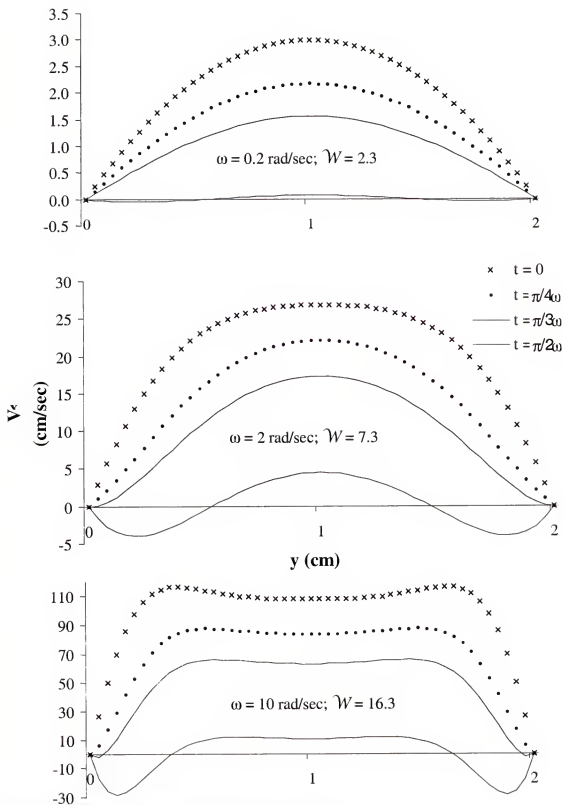


Figure 2.4 Velocity profiles at three different frequencies.

more rapidly than the slower diffusing species. Although some of the slower diffuser may reach the boundary, on average more of the slower diffusing species resides near the faster moving core than the faster diffusing species causing it to transport more than the faster diffuser.

Looking at Fig. 2.3, we see that the ratio of the total mass transport of the slow diffuser to the fast diffuser becomes unity as the frequency of oscillation increases and then proceeds to decrease toward a maximum. Now as the frequency increases even more, it takes less time to complete a cycle and neither species has enough time to reach the boundary of the fluid before the flow is reversed on the next part of the cycle. Over this frequency range, the faster diffuser has the advantage of being able to move in and out of the core taking advantage of the forced flow without getting bogged down in the slow flow region. Although the slower diffuser is able to move somewhat, its lower mobility and the higher frequency of the flow do not allow as much of it to move in and out of the core as the faster diffuser does. The faster diffusing species will therefore once again begin to overtake the slower diffuser as it travels down the tube in a zigzag fashion.

Another minimum is achieved as the frequency is made greater than before and the mass transfer of the slower species once again increases more rapidly than the faster diffuser. In this region of high frequency the “wiggles” in the velocity profile begin to affect the total mass transfer of both species. The maximum velocity is no longer in the center of the gap, and a region of reverse flow appears at certain times during the cycle. Fig. 2.4 gives an idea of the velocity profiles for various times in a cycle of different frequencies.³ The faster diffuser moves a greater distance than the slower diffuser, and as

³ These flow profiles have the same qualitative form as those found by Uchida [14] for pulsating flow in a circular pipe.

it moves into the region of reverse flow, its overall mass transfer is hindered. The slower diffuser once again stays near the core, and as not as much of it has time to move in and out of the core, not as much of it moves into the region of reverse flow either. This once again gives the slower moving species the advantage over the faster diffuser.

Returning to Fig. 2.3, it is clear that the crossovers occur at different frequencies for the different systems. In fact, depending on the properties of the system, all of the crossovers may or may not occur. On first examination, it is apparent that the diffusion coefficients contribute to the crossovers and the maximum in Fig. 2.3. The ratio of diffusion coefficients is greatest for the He-CO₂ system and smallest for the He-X system as is evidenced in Fig. 2.3 at zero frequency. Considering first the He-CO₂ system, CO₂ has a large difference in diffusivity to initially overcome, because it remains much nearer to the core compared to Helium as was previously explained. Consequently, its transport will overtake Helium and the ratio of the total mass transport will actually have a lower minimum than the other two systems. Because of the large disparity between the diffusion coefficients of CO₂ and Helium, the slower diffuser in this system will have a higher mass transfer compared to the faster diffuser than in the corresponding situation in the other two systems. For systems with large diffusivity ratios such as the He-CO₂ and He-CH₄ systems, a second crossover will occur as the frequency increases only if the faster diffuser is able to make up the ground it lost before the reverse flow produced by even higher frequencies causes it to decrease again. However, the He-X system has a small diffusivity ratio, and although the mass transfer of Helium initially begins to overtake the mass transfer of species X after the maximum occurs in the ratio of the total mass transfer, it is not enough to overtake species X before the reverse flow hinders the Helium. For this reason, only one crossover exists for the He-X binary system. As the

reverse flow becomes more prominent, it could possibly lead to a third crossover as evident in the He-CH₄ system. The third crossover therefore occurs if the final increase after the second crossover is enough to allow the slower diffuser to once again overtake the faster diffuser.⁴ The second and third crossovers are related to the change in the flow profiles with frequency and can be seen by manipulating the values of the diffusion coefficient or the values of the kinematic viscosity.

An increase in the kinematic viscosity causes the fluid to respond to input changes more readily. As an easily seen example, the expression for V_x in the physical situation of an oscillating wall in an infinitely deep channel given in Chapter 1 is

$$V_x = \frac{1}{2} A \omega e^{-y\sqrt{\omega/2\nu}} \cos\left(y\sqrt{\omega/2\nu} - \omega t\right) \quad (1.10)$$

It can be observed that large values of ν damp out the phase lag that is the primary cause for “wiggles” in the flow. Indeed, as ν approaches infinity, there is no phase lag and no “wiggles” because the response of the fluid velocity to the plate velocity is instantaneous. Consequently, for finite kinematic viscosity, the “wiggles” that we expect to see are delayed to larger frequencies as the value of ν is increased as is seen in Fig. 2.5 comparing the velocity profile at 10 rad/sec for three different kinematic viscosities. This gives a larger range of frequencies where “wiggles” are absent and where the transport of the faster diffuser increases more than the slower diffuser as seen in Fig. 2.6. This increase causes the local minimum in the ratio of mass transfer to decrease with an

⁴ On a practical note, the third crossover occurs at a value of ω of roughly 14 rad/sec. In order to avoid end and entrance effects for a plate spacing of 2 cm, amplitude of 20 cm, and kinematic viscosity of 0.15 cm²/sec, it is estimated that this requires a distance of around 1.2 meters or more between the reservoirs. Smaller plate spacings would allow us to get away with smaller distances between reservoirs, but this, as imagined, might come at the cost of losing a crossover.

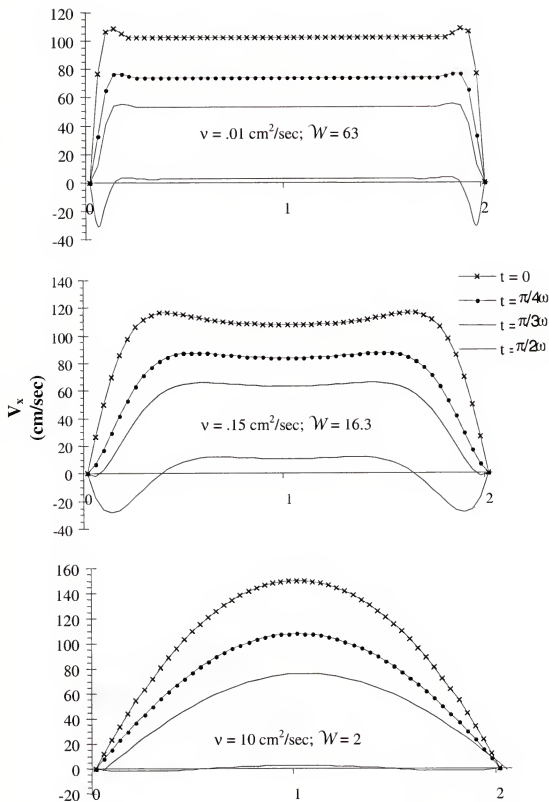


Figure 2.5 Velocity profiles for three different kinematic viscosities ($\omega = 10$ rad/sec).

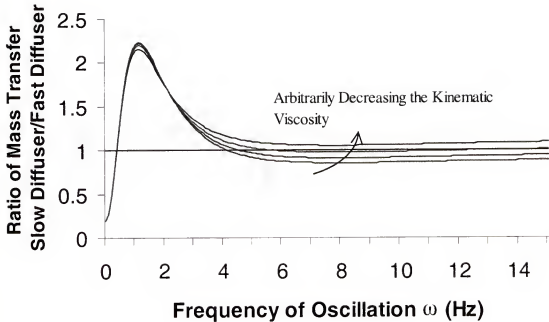


Figure 2.6 Separation ratios for a He – CH₄ system with different kinematic viscosities of the carrier fluid.

increase in ν . The third crossover may not therefore occur for reasonable frequencies for large kinematic viscosities. It can also be noted, but only briefly mentioned here, that changing the value of the width between the plates will also change the number of crossovers that occur for the different systems. It can be readily seen that changing the width will change the velocity profiles between the plates and thus affecting the mass transfer for both species in much the same way as varying the kinematic viscosity.

The same phenomenon also occurs in a liquid medium as evidenced in Fig. 2.7. For liquids, the diffusion coefficients are very small compared to gases, but the kinematic viscosity is only a little smaller. Because of the very small diffusion coefficients, the overall mass transfer of the binary system is much smaller than in gases as expected. It is also seen that the crossovers occur at much smaller frequencies than for gases. They occur at such small frequencies that the crossovers may be undetectable within the

accuracy of most instruments. The behavior of the ratios of mass transfer acts exactly the same as in gases except on a smaller scale, as expected. Here too, the crossovers can be manipulated by varying the kinematic viscosity, the diffusion coefficients, or the height.

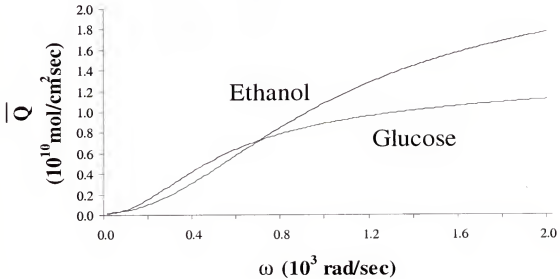


Figure 2.7 Total mass transfer versus frequency for ethanol and glucose in a water carrier.

As has been noted by previous authors [8-10,16-18], pulsatile flow can be used to achieve a reasonable separation instead of using other traditional methods. It only requires mechanical work and produces no net flow from one reservoir to another. And, as can be seen in Fig. 2.2 and Fig. 2.3, there exists a trade off between mass transfer and separation achieved. With an increase in frequency, the separation will decrease while the total mass transfer will increase. There also exist frequencies where no separation will exist at all as the total mass transfer for each species is the same. It is finally noted that no crossovers need occur if the maximum velocity is small enough (small piston amplitudes). Under such circumstances, the diffusive mass transport far exceeds any convective mass transport. This comes as no surprise.

2.3 Endnote 1: Pressure Driven Versus Boundary Driven Periodic Systems

Now, it turns out that there are several ways to induce pulsatile flow. In addition to an imposed oscillating pressure drop studied in this chapter, the tube walls of the channel may also be oscillated as evidenced in the introductory chapter for an infinite fluid. In what follows, an analysis will be provided to show the differences and to point out the similarities between a configuration driven by an oscillating piston and one by oscillating boundaries. The mass transfer will be calculated and shown to vary between the two cases. Another important parameter that will be calculated for each periodic configuration will be the power required to drive the flow. This will then allow a comparison of the power required for the mass transfer produced by each method.

2.3.1 Pressure driven case revisited

In the previous section, the mass transfer was given in unscaled quantities in order to understand the physics behind the phenomena produced by periodic flow. Now that the physics has been explained, the mass transfer for a pressure driven system will be scaled resulting in the Womersley number, the Schmidt number, and other dimensionless parameters. Again, the Womersley number is defined as

$$W \equiv h \sqrt{\frac{\omega}{2\nu}} \quad (2.13)$$

and the Schmidt number is

$$Sc \equiv \frac{\nu}{D} \quad (2.14)$$

Note that the Womersley number has a $\sqrt{2}$ in the denominator; therefore, it may be different than the definition of the Womersley number of previous authors.

In scaled quantities, the velocity profile in pressure driven flow now is given by

$$\hat{V}_x = \frac{\frac{1}{4} A \omega \hat{W}}{(2 - \hat{W})e^{\hat{W}} + (2 + \hat{W})e^{-\hat{W}} - 4} \left[(1 - e^{-\hat{W}})e^{\hat{W}y^*} + (e^{\hat{W}} - 1)e^{-\hat{W}y^*} + e^{-\hat{W}} - e^{\hat{W}} \right] \quad (2.15)$$

$$\tilde{V}_x = \frac{\frac{1}{4} A \omega \tilde{W}}{(\tilde{W} - 2)e^{\tilde{W}} - (\tilde{W} + 2)e^{-\tilde{W}} + 4} \left[(e^{-\tilde{W}} - 1)e^{\tilde{W}y^*} + (1 - e^{\tilde{W}})e^{-\tilde{W}y^*} + e^{\tilde{W}} - e^{-\tilde{W}} \right] \quad (2.16)$$

where $y^* = \frac{y}{h}$, $\hat{W} = (i+1)W$, and $\tilde{W} = (i-1)W$. The expression for $\hat{c}$ is given by

$$\hat{c} = \frac{\frac{A}{4} i(c_2 - c_1) \hat{W} S c}{(S c - 1) \left[(2 - \hat{W})e^{\hat{W}} + (2 + \hat{W})e^{-\hat{W}} - 4 \right]} \times \left[\frac{(1 - e^{-\hat{W}})(e^{\hat{W}} - e^{\hat{W}\sqrt{S c}}) + (e^{\hat{W}} - 1)(e^{\hat{W}\sqrt{S c}} - e^{-\hat{W}})}{e^{-\hat{W}\sqrt{S c}} - e^{\hat{W}\sqrt{S c}}} S c^{-1/2} e^{-\hat{W}\sqrt{S c}y^*} \right. \\ \left. + \frac{(1 - e^{-\hat{W}})(e^{\hat{W}} - e^{-\hat{W}\sqrt{S c}}) + (e^{\hat{W}} - 1)(e^{-\hat{W}\sqrt{S c}} - e^{-\hat{W}})}{e^{-\hat{W}\sqrt{S c}} - e^{\hat{W}\sqrt{S c}}} S c^{-1/2} e^{\hat{W}\sqrt{S c}y^*} \right. \\ \left. + (e^{\hat{W}} - 1)e^{-\hat{W}y^*} + (1 - e^{-\hat{W}})e^{\hat{W}y^*} + (e^{\hat{W}} - e^{-\hat{W}})(S c^{-1} - 1) \right] \quad (2.17)$$

As noted earlier, for gases, Sc is of the order of unity which indicates viscous and diffusive effects are equally important, and for liquids, $Sc \sim 1000$ showing that viscous effects are significantly more important than diffusion. In deriving the total mass transfer, it will again be noted that

$$\tilde{V}_x = \frac{1}{4} A \omega (\psi_1 e^{\tilde{W}y^*} + \psi_2 e^{-\tilde{W}y^*} + \psi_3) \quad (2.18)$$

$$\hat{c} = \frac{1}{4} A \frac{(c_2 - c_1)}{L} (\phi_1 e^{-\hat{W}\sqrt{S c}y^*} + \phi_2 e^{\hat{W}\sqrt{S c}y^*} + \phi_3 e^{-\hat{W}y^*} + \phi_4 e^{\hat{W}y^*} + \phi_5) \quad (2.19)$$

where the ψ 's and ϕ 's are all constants depending on the parameters of the system (D , ω , ν , and h) written this time in terms of the Womersley and Schmidt numbers. The expressions for the ψ 's and ϕ 's can be derived upon inspection of $\tilde{V}_x$ and $\hat{c}$ from the above equations. The final expression for the total mass transfer is then

$$\bar{Q} = -\frac{D(c_2 - c_1)}{L} + \frac{A^2 \omega (c_2 - c_1)}{8L} \Re \left[\begin{aligned} & \frac{\psi_1 \phi_1 (e^{\tilde{W} - \hat{W} \sqrt{Sc}} - 1) - \psi_2 \phi_2 (e^{\hat{W} \sqrt{Sc} - \tilde{W}} - 1)}{\tilde{W} - \hat{W} \sqrt{Sc}} \\ & + \frac{\psi_1 \phi_2 (e^{\tilde{W} + \hat{W} \sqrt{Sc}} - 1) - \psi_2 \phi_1 (e^{-(\tilde{W} + \hat{W} \sqrt{Sc})} - 1)}{\tilde{W} + \hat{W} \sqrt{Sc}} \\ & + \frac{\psi_1 \phi_3 (e^{\tilde{W} - \hat{W}} - 1) - \psi_2 \phi_4 (e^{\hat{W} - \tilde{W}} - 1)}{\tilde{W} - \hat{W}} \\ & + \frac{\psi_1 \phi_4 (e^{\tilde{W} + \hat{W}} - 1) - \psi_2 \phi_3 (e^{-(\tilde{W} + \hat{W})} - 1)}{\tilde{W} + \hat{W}} \\ & + \frac{\psi_1 \phi_5 (e^{\tilde{W}} - 1) - \psi_2 \phi_5 (e^{-\tilde{W}} - 1)}{\tilde{W}} \\ & + \frac{\psi_3 \phi_2 (e^{\hat{W} \sqrt{Sc}} - 1) - \psi_3 \phi_1 (e^{\hat{W} \sqrt{Sc}} - 1)}{\hat{W} \sqrt{Sc}} \\ & + \frac{\psi_3 \phi_4 (e^{\hat{W}} - 1) - \psi_3 \phi_3 (e^{-\hat{W}} - 1)}{\hat{W}} + \psi_3 \phi_5 \end{aligned} \right] \quad (2.20)$$

In order to non-dimensionalize the above equation of total mass transfer, it will first be normalized with respect to the diffusive mass transfer

$$Q_{diff} = -\frac{D(c_2 - c_1)}{L} \quad (2.21)$$

Notice that the term within the $\Re$ is dimensionless and is only a function of Womersley and Schmidt numbers. This leaves the term in front of the $\Re$, upon normalization, to be

$$-\frac{A^2 \omega}{8D} \quad (2.22)$$

Jaeger et al. [10] scale their effective diffusion coefficient, D_{eff} , by $A^2 \omega$ in order to make it dimensionless. This implies that in their plots of the scaled effective diffusion coefficient versus Womersley number that the frequency and kinematic viscosity are held fixed and the Womersley number is varied through the gap width, h (or through the radius of the tube in a cylindrical geometry). Another way is to hold the width fixed and vary the Womersley number by varying the frequency, ω . This would give

$$-\frac{A^2\omega}{8D} = -\frac{1}{8} \frac{A^2}{h^2} W^2 Sc \quad (2.23)$$

This difference in scaling is the reason why Jaeger et al. achieve a maximum in their figures showing the effective diffusion coefficient versus Womersley number and why the figures in this thesis show a continuous monotonic increase in the mass transfer for increasing Womersley number like those found by Harris and Goren [3]. Using the above expression, the dimensionless form of the mass transfer for a pressure driven system is

$$\frac{\bar{Q}}{D(c_1 - c_2)} = 1 - \frac{A^2 W^2 Sc}{4h^2} \Re \left[\begin{aligned} & \frac{\psi_1 \varphi_1 (e^{\tilde{W} - \hat{W} \sqrt{Sc}} - 1) - \psi_2 \varphi_2 (e^{\hat{W} \sqrt{Sc} - \tilde{W}} - 1)}{\tilde{W} - \hat{W} \sqrt{Sc}} \\ & + \frac{\psi_1 \varphi_2 (e^{\tilde{W} + \hat{W} \sqrt{Sc}} - 1) - \psi_2 \varphi_1 (e^{-(\tilde{W} + \hat{W} \sqrt{Sc})} - 1)}{\tilde{W} + \hat{W} \sqrt{Sc}} \\ & + \frac{\psi_1 \varphi_3 (e^{\tilde{W} - \hat{W}} - 1) - \psi_2 \varphi_4 (e^{\hat{W} - \tilde{W}} - 1)}{\tilde{W} - \hat{W}} \\ & + \frac{\psi_1 \varphi_4 (e^{\tilde{W} + \hat{W}} - 1) - \psi_2 \varphi_3 (e^{-(\tilde{W} + \hat{W})} - 1)}{\tilde{W} + \hat{W}} \\ & + \frac{\psi_1 \varphi_5 (e^{\tilde{W}} - 1) - \psi_2 \varphi_5 (e^{-\tilde{W}} - 1)}{\tilde{W}} \\ & + \frac{\psi_3 \varphi_2 (e^{\hat{W} \sqrt{Sc}} - 1) - \psi_3 \varphi_1 (e^{\hat{W} \sqrt{Sc}} - 1)}{\hat{W} \sqrt{Sc}} \\ & + \frac{\psi_3 \varphi_4 (e^{\hat{W}} - 1) - \psi_3 \varphi_3 (e^{-\hat{W}} - 1)}{\hat{W}} + \psi_3 \varphi_5 \end{aligned} \right] \quad (2.24)$$

Note that in this case, $c_1 > c_2$ so that the normalized mass transfer is a positive quantity.

This dimensionless form of the mass transfer is now only dependent on the Womersley number (W), the Schmidt Number (Sc), and the ratio of the piston amplitude to the distance between the plates (A/h).

Before moving to the analysis of the power required to drive the flow, a brief digression is made on the pressure used to impose the periodic flow. Consider the

original equation of motion for this configuration given by Equation 2.2. Suppose that the pressure oscillations are generated in some arbitrary way and not necessarily by only moving piston heads. The pressure drop can still be written as the sum of time-complex exponentials as

$$\frac{1}{\mu} \frac{\Delta P}{L} = \hat{P}(A, \omega, \nu, h) e^{i\omega t} + \tilde{P}(A, \omega, \nu, h) e^{-i\omega t} \quad (2.25)$$

where $\hat{P}$ and $\tilde{P}$ are functions of the parameters of the problem and are conjugate to each other. The solution for $\hat{V}_x$ for a general pressure drop is then

$$\hat{V}_x = -\frac{\frac{1}{2} \hat{P} h^2 i}{W^2 (e^{\hat{W}} - e^{-\hat{W}})} \left[(1 - e^{-\hat{W}}) e^{\hat{W} y^*} + (e^{-\hat{W}} - 1) e^{-\hat{W} y^*} + e^{-\hat{W}} - e^{\hat{W}} \right] \quad (2.26)$$

Note that $\hat{V}_x$ is homogeneously dependent on the pressure term, $\hat{P}$. This is also true of its conjugate, $\tilde{V}_x$, as it depends homogeneously on $\tilde{P}$. As the velocity is the inhomogeneous term in the species conservation equation, $\hat{c}$ and $\tilde{c}$ will also be homogeneous in $\hat{P}$ and $\tilde{P}$ respectively. This will make the convective mass transfer quadratic in pressure, i.e., $|\hat{P}|^2$, and when the pressure drop is induced by oscillating piston heads as described earlier, it will be homogeneous in A^2 . This realization will become important in comparing the pressure driven method to the boundary driven later on.

Returning now to the piston driven configuration, the power can be found by taking the dot product of the velocity vector with the equation of motion and then integrating over the entire fluid region. This shows that the energy required to drive the fluid is equal to the kinetic energy plus the frictional heat dissipated by the system. This

is only the power required to move the fluid, and it does not account for the inertia of the piston itself. For the pressure driven case

$$Power = \frac{\Delta P}{L} \int_0^h V_x dy \quad (2.27)$$

The time averaged power, $\overline{Power}$, over one complete cycle is

$$\overline{Power} = 2\Re \left(\frac{\Delta \tilde{P}}{L} \int_0^h \hat{V}_x dy \right) \quad (2.28)$$

or

$$\overline{Power} = \frac{\mu A^2 \omega^2}{8h} \Re \left[\frac{\hat{W}^3 (e^{\hat{W}} - e^{-\hat{W}})}{(2 - \hat{W})e^{\hat{W}} + (2 + \hat{W})e^{-\hat{W}} - 4} \right] \quad (2.29)$$

Because the geometry is two dimensional and infinite in extent, the time-averaged power calculated is essentially a power per area of fluid. The dimensionless time-averaged power becomes

$$\frac{\overline{Power}}{\mu h \omega^2} = \frac{1}{2} \frac{A^2}{h^2} \Re \left[\frac{\hat{W}^3 (e^{\hat{W}} - e^{-\hat{W}})}{(2 - \hat{W})e^{\hat{W}} + (2 + \hat{W})e^{-\hat{W}} - 4} \right] \quad (2.30)$$

Note from the above equation that the time-averaged power is also quadratic function of the pressure in the same way as the convective mass transfer previously discussed.

2.3.2 Boundary Driven Case

Moving on to the case of periodic flow induced by the oscillation of the walls we see that the power and mass transfer for this case are obtained in a manner similar to the pressure driven case. In this configuration, imagine Fig. 2.1 with the pistons removed and the walls of the channel oscillating in phase as $V_{wall} = \frac{1}{2} A \omega \cos(\omega t)$. Once again, assuming the fluid to be incompressible, Newtonian, with a kinematic viscosity ν , and the flow to be laminar, the x -component of the equations of motion for this system is

$$\frac{\partial V_x}{\partial t} = v \frac{\partial^2 V_x}{\partial y^2} \quad (2.31)$$

Observe here that a pressure gradient neither drives nor is induced by the flow. $\hat{V}_x$

becomes

$$\hat{V}_x = \frac{1}{4} A \omega \left[\frac{e^{-\tilde{w}} e^{\tilde{w}_{y^*}} + e^{-\tilde{w}_{y^*}}}{1 + e^{-\tilde{w}}} \right] \quad (2.32)$$

and $\tilde{V}_x$ is

$$\tilde{V}_x = \frac{1}{4} A \omega \left[\frac{e^{\tilde{w}_{y^*}} + e^{-\tilde{w}} e^{-\tilde{w}_{y^*}}}{1 + e^{-\tilde{w}}} \right] \quad (2.33)$$

$\hat{c}$ for the boundary driven system is

$$\hat{c} = \frac{\frac{1}{4} Ai (c_2 - c_1) Sc^{1/2} \left(1 - e^{-\tilde{w}} \right) \left[e^{-\tilde{w} \sqrt{Sc} y^*} + e^{-\tilde{w} \sqrt{Sc}} e^{\tilde{w} \sqrt{Sc} y^*} \right]}{L(Sc - 1) \left(1 + e^{-\tilde{w}} \right) \left(e^{-\tilde{w} \sqrt{Sc}} - 1 \right)} + \frac{\frac{1}{4} Ai (c_2 - c_1) Sc}{L(Sc - 1) \left(1 + e^{-\tilde{w}} \right)} \left[e^{-\tilde{w}} e^{\tilde{w}_{y^*}} + e^{-\tilde{w}_{y^*}} \right] \quad (2.34)$$

The total time- and space-averaged mass transfer have the same general form as in the pressure driven case. Again, defining the velocity and concentration as

$$\tilde{V}_x = \frac{1}{4} A \omega \left(\beta_1 e^{\tilde{w}_{y^*}} + \beta_2 e^{-\tilde{w}_{y^*}} \right) \quad (2.35)$$

$$\hat{c} = \frac{\frac{1}{4} A (c_2 - c_1)}{L} \left(\eta_1 e^{-\tilde{w} \sqrt{Sc} y^*} + \eta_2 e^{\tilde{w} \sqrt{Sc} y^*} + \eta_3 e^{\tilde{w}_{y^*}} + \eta_4 e^{-\tilde{w}_{y^*}} \right) \quad (2.36)$$

where the β 's and the η 's can be determined by inspection from Equation 2.33 and

Equation 2.34. The mass transfer in dimensionless form becomes

$$\frac{\bar{Q}}{L} = 1 - \frac{A^2 W^2 Sc}{4h^2} \Re \left[\begin{aligned} & \frac{\beta_1 \eta_1 \left(e^{\tilde{W} - \hat{W} \sqrt{Sc}} - 1 \right) - \beta_2 \eta_2 \left(e^{\hat{W} \sqrt{Sc} - \tilde{W}} - 1 \right)}{\tilde{W} - \hat{W} \sqrt{Sc}} \\ & + \frac{\beta_1 \eta_2 \left(e^{\tilde{W} + \hat{W} \sqrt{Sc}} - 1 \right) - \beta_2 \eta_1 \left(e^{-(\tilde{W} + \hat{W} \sqrt{Sc})} - 1 \right)}{\tilde{W} + \hat{W} \sqrt{Sc}} \\ & + \frac{\beta_1 \eta_3 \left(e^{\tilde{W} + \hat{W}} - 1 \right) - \beta_2 \eta_4 \left(e^{-(\tilde{W} + \hat{W})} - 1 \right)}{\tilde{W} + \hat{W}} \\ & + \frac{\beta_1 \eta_4 \left(e^{\tilde{W} - \hat{W}} - 1 \right) - \beta_2 \eta_3 \left(e^{\hat{W} - \tilde{W}} - 1 \right)}{\tilde{W} - \hat{W}} \end{aligned} \right] \quad (2.37)$$

The power required to drive a boundary driven system is also the sum of the kinetic energy and the frictional heat dissipated by it. This is equal to the product of the velocity and the shear stress at the boundaries, and is therefore

$$Power = -2\mu V_x \left. \frac{\partial V_x}{\partial y} \right|_{y=0} \quad (2.38)$$

After taking the time average over one cycle, the total dimensionless power over a cycle is then

$$\frac{\overline{Power}}{\mu h \omega^2} = \frac{1}{4} \frac{A^2}{h^2} \Re \left[\frac{\hat{W} (e^{-\hat{W}} - 1)}{1 + e^{-\hat{W}}} \right] \quad (2.39)$$

Notice that here too, the convective part of the mass transfer and the time-averaged power are both homogeneous in the square of the amplitude of the wall displacement.

2.3.3 A Comparison of both Pulsatile Methods and Some Results

Fig. 2.8 shows the *convective* mass transfer increasing with Womersley number and is also the same type of result seen by Harris and Goren [3]. Note that the comparison is made for the same value of “A” where A represents the peak-to-peak amplitude of the piston head in the one case and the peak-to-peak displacement of the

tube walls in the other. It is seen that convective mass transfer of a single species for both methods under the same conditions and given parameters is greater for the pressure

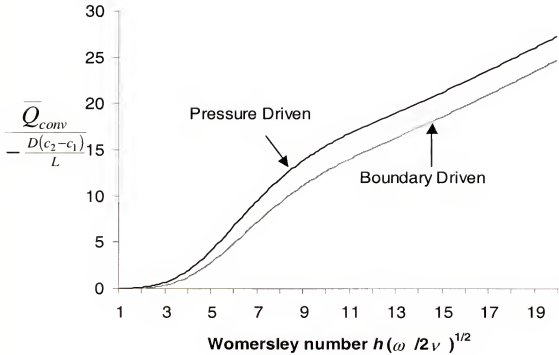


Figure 2.8 Scaled convective mass transfer versus Womersley number for a pressure driven and boundary driven configuration.

driven method for all Womersley numbers than the boundary driven method. This is conceivably due to the fact that in a boundary driven configuration, most of the mass transfer takes place in a small viscous boundary layer near the surfaces of the plates where most of the fluid is moving. However, for the pressure driven method, more fluid moves between the two plates due to the imposed pressure drop which gives rise to a larger region for mass transfer to take place. As more fluid moves, the drawback of the pressure driven case is that more power is required to drive it than a boundary driven configuration. The ratio of convective mass transfer to power required for both methods are then calculated as this gives a reasonable comparison between both systems; the ratio

being independent of amplitude in both the pressure and boundary driven cases. This is important because the value of the amplitude of the piston stroke for the pressure driven case is not necessarily the amplitude in the boundary driven case. By eliminating the amplitude in the convective mass transfer to power ratio, the problem of dealing with two different amplitudes is no longer an issue. *It was found that the values for this ratio for all Womersley numbers is identical in both cases.* This shows that there is a direct relationship between a pressure driven and boundary driven configuration which can be further demonstrated by a moving frame calculation for the boundary driven method.

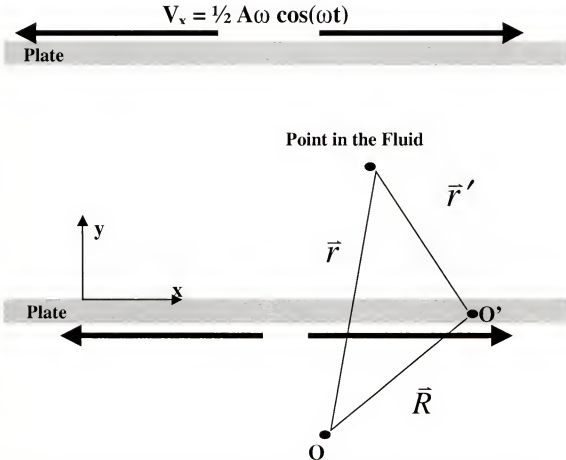


Figure 2.9 Moving frame picture of a point in the fluid from a fixed observer and a moving observer oscillating with the plates.

As the mass transfer of a system is a scalar quantity, it is independent of the observer's point of reference in relation to the system. Consider the point of view of two different observers: one fixed observer (O) and one observer sitting on the moving plate (O') in the boundary driven case. In Fig. 2.9, the relation between O and O' is then

$\vec{R} = \frac{1}{2} A \cos(\omega t) \vec{i}$ and by vector addition $\vec{R} + \vec{r}' = \vec{r}$ where $\vec{r}$ and $\vec{r}'$ are the position

vectors of a point as viewed by the observers O and O' respectively. From this, the x -component of the equations of motion for the moving frame calculation is then

$$\rho \frac{\partial V'_x}{\partial t} = \mu \frac{\partial^2 V'_x}{\partial y^2} + \frac{1}{2} A \omega^2 \cos(\omega t) \quad (2.40)$$

subject to

$$V'_x = 0 \quad \text{at } y=0 \text{ and } y=h \quad (2.41, 2.42)$$

Here, V'_x is the velocity measured by O' . When comparing this to the equation of motion for the fixed observer boundary driven case, Equation 2.31, an extra term is apparent in the equation. This is similar to a time-dependent pressure term apparent in the equation of motion for the pressure driven case making

$$\frac{\Delta P}{L} = \frac{1}{2} A \omega^2 \cos(\omega t) \quad (2.43)$$

which is once again dependent on the amplitude of oscillation imposed upon the system.

One can imagine sitting on the moving plate and observing the fluid oscillating back and forth. Since the observer can not determine that the plate is moving, the observer would assume only a periodic pressure gradient could be causing the fluid to oscillate. This time-dependent "pressure gradient" is observed in the equation of motion for the moving frame. As power required to drive the system would be the same regardless of the

reference point of either observer, the ratio of convective mass transfer to power will be the same for the fixed and moving frame since it is exactly the same system.

Although it may seem surprising, at first, that periodic flow driven by an oscillating piston and periodic flow in a boundary driven configuration will give exactly the same convective mass transfer to power ratio, this turns out to also be true for any generalized oscillating pressure drop that can be written in the form of Equation 2.25. As noted earlier, the convective mass transfer is quadratic in pressure as also the power required to drive this configuration, so their ratio must be independent of the pressure drop and its origin. Further, as the boundary driven method can be written as an oscillating pressure in a moving frame of reference, its convective mass transfer to power ratio will be the same as any general pressure driven problem.

Next we consider the case where the diffusive mass transfer *and* the convective mass transfer are both taken into account in the calculation of the *total* mass transfer. This is important at low frequencies as the diffusive mass transfer then plays a part in the total mass transfer of the system. Now, the differences between pressure driven and boundary driven configurations become more apparent. The diffusive mass transfer does not depend on the amplitude driving the system or the Womersley number, and it is the same for both configurations provided the same axial concentration gradient, diffusion coefficient, and length between reservoirs are assumed. The ratio of total mass transfer, which is again the sum of diffusive and convective mass transfer, to the power applied will therefore be different for various Womersley numbers as the power increases for increasing Womersley numbers. If the parameters for the boundary driven and pressure driven cases are the same, the total mass transfer per power applied for the boundary driven case will be higher than the pressure driven case at low Womersley numbers

because the power required to drive a pressure driven system is higher than a boundary driven system. This is seen in Fig. 2.10. Of course, the periodic pressure gradient that

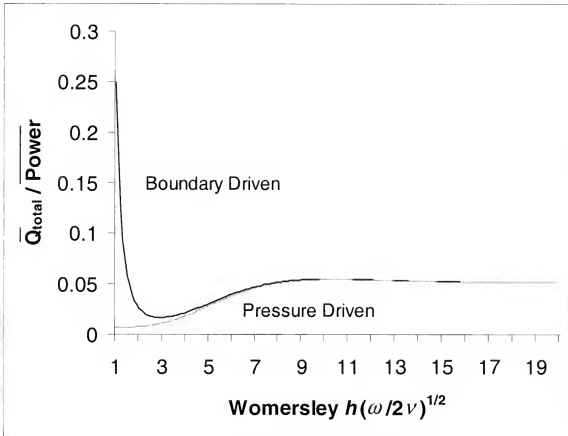


Figure 2.10 Scaled total mass transfer per power versus Womersley number for a boundary and pressure driven configuration.

will give the same ratio of total mass transfer to power as the boundary driven case will be the pressure drop derived from the moving frame calculation.

This then leads to the question of what the value of Womersley number will be for the ratio of total mass transfer per power for both methods to be equal to each other. Fig. 2.10 demonstrates that the boundary driven and pressure driven configurations eventually asymptote toward the same value with increasing Womersley number as expected. For a mixture of dilute Helium ($Sc = 0.21$) in a Nitrogen carrier, the point where the total mass transfer is within 0.1% of each other corresponds to a Womersley

number of 39.3. For reference, a system consisting of dilute CO_2 ($Sc = 0.91$) in a Nitrogen carrier under the same conditions has a corresponding Womersley number of 17.6. This suggests that the system with the higher diffusion coefficient (Helium) requires a higher Womersley number in order for the convective mass transfer to dominate over pure molecular diffusion. Because the species with the lower diffusion coefficient (CO_2) resides nearer to the faster moving core of the fluid over one cycle, the slower diffusing CO_2 takes advantage of the periodic convection more effectively than the faster diffusing Helium. This allows the convective mass transfer to dominate the process at a smaller Womersley number.

2.4 Endnote 2: Justification of the Conjecture that Concentration, $c(x, r, t)$, is

$$\frac{\Delta c}{L} x + c^*(r, t)$$

By supposing that the concentration is the sum of an axial part plus a time and radial dependent part is key to solving the periodic flow problems by analytical means. Harris and Goren [3] were one of the earliest to make this approximation. They believed that there was a mean part of the concentration that varies linearly with axial distance in saying “Since the gradient of the mean concentration is constant, the fluctuations in concentration brought about by pulsation are independent of axial distance. This is because events at any two cross sections of the tube are identical except for the absolute value of the concentration, as the amplitude of the concentration fluctuations is proportional to the concentration gradient and not to the concentration itself. When this is the case, second and higher harmonics are not introduced.” Watson [4] used the same assumption in his paper and further concluded that if a steady flow was superimposed upon periodic flow in a pipe, Taylor’s [2] dispersion result would be recovered as the frequency of oscillation became zero. This assessment points to Taylor as the source of

the concentration approximation, and so it is Taylor's dispersion problem that is first discussed.

2.4.1 Taylor Dispersion

In Taylor's steady flow problem, the dispersion of a drop of fluorescent solution in a stream of slow moving water in a tube was predicted. The governing equation to model dispersion is the species conservation equation with a steady flow in a tube. It is

$$\frac{\partial c}{\partial t} + u_0 \left(1 - \frac{r^2}{a^2} \right) \frac{\partial c}{\partial x} = D \left(\frac{\partial^2 c}{\partial r^2} + \frac{1}{r} \frac{\partial c}{\partial r} + \frac{\partial^2 c}{\partial x^2} \right) \quad (2.44)$$

where u_0 is the maximum velocity in a tube of radius a with impermeable walls. In order to find the concentration field analytically, Taylor assumes that the molecular diffusion in the longitudinal direction is small compared to the radial diffusion. Next, upon placing the observer on a frame moving at the mean velocity of the flow, or $\frac{1}{2} u_0$, only radial variations of the concentration are seen, and the concentration is independent of time and axial changes. Neglecting pure molecular diffusion, the convective mass transfer is then

$$Q = - \frac{\pi a^4 u_0^2}{192D} \frac{\partial \bar{c}}{\partial x} \quad (2.45)$$

where x is the axial coordinate and $\bar{c}$ is its mean concentration. According to Taylor, in the moving frame, the variation of concentration along the axial direction is at most a function of only x and only the *mean* concentration is involved. He calculates an effective diffusion coefficient to be

$$D_{eff} = \frac{a^2 u_0^2}{192D} \quad (2.46)$$

which is the famous Taylor dispersion result for steady flow in a tube that shows that dispersion is inversely proportional to the molecular diffusion coefficient.

2.4.2 Aris' Moment Verification

Now, Taylor's calculation was a result of the approximations that he perceived on physical grounds. Aris [19], a few years later, revisited this problem and used the method of moments to solve for the dispersion of solute in a tube in order to see if he arrived at the same conclusion thereby justifying the approximations of Taylor. Moments are taken of Equation 2.42 where the n^{th} moment of concentration is defined as

$$c_n = \int_{-\infty}^{+\infty} x^n c dx \quad (2.47)$$

The first three moment equations ($n = 0, 1, 2$) for the concentration then become

$$\begin{aligned} \frac{\partial c_0}{\partial t} &= \frac{D}{r} \frac{\partial}{\partial r} \left(r \frac{\partial c_0}{\partial r} \right) \\ \frac{\partial c_1}{\partial t} &= \frac{D}{r} \frac{\partial}{\partial r} \left(r \frac{\partial c_1}{\partial r} \right) + V_x c_0 \\ \frac{\partial c_2}{\partial t} &= \frac{D}{r} \frac{\partial}{\partial r} \left(r \frac{\partial c_2}{\partial r} \right) + 2V_x c_1 + 2Dc_0 \end{aligned} \quad (2.48, 2.49, 2.50)$$

If one were to examine the mean concentration in the tube that has already been spatially averaged over a cross section, the conservation equation becomes

$$\frac{\partial \bar{c}}{\partial t} + V_{eff} \frac{\partial \bar{c}}{\partial x} = D_{eff} \frac{\partial^2 \bar{c}}{\partial x^2} \quad (2.51)$$

where V_{eff} is the effective velocity and D_{eff} is the effective diffusivity. Moments can also be defined for the mean concentration so that

$$\bar{c}_n = \int_{-\infty}^{+\infty} x^n \bar{c} dx \quad (2.52)$$

to give the first three corresponding moment equations

$$\begin{aligned}
\frac{\partial \bar{c}_0}{\partial t} &= 0 \\
\frac{\partial \bar{c}_1}{\partial t} &= V_{eff} \bar{c}_0 \\
\frac{\partial \bar{c}_2}{\partial t} &= 2V_{eff} \bar{c}_1 + 2D_{eff} \bar{c}_0
\end{aligned} \tag{2.53, 2.54, 2.55}$$

The solution to Equations 2.48, 2.49, and 2.50 can be calculated and related to the mean concentration by the definition that the mean concentration is the spatial average of the concentration field, or

$$\bar{c}_n = \frac{\int_0^{2\pi} \int_0^a c_n r dr d\theta}{\int_0^{2\pi} \int_0^a r dr d\theta} \tag{2.56}$$

This relationship can be used to determine the values for V_{eff} and D_{eff} . Aris, and later rederived in lecture notes by Johns [20] using eigenfunction expansions, found that V_{eff} is simply V_x , or the steady flow velocity in the tube, and D_{eff} is exactly the result found by Taylor. This proves that Taylor's approximations were indeed justified. Before continuing to the periodic case, a derivation similar to the one given for a circular geometry is reproduced for steady flow in a channel, showing its general applicability.

2.4.3 Dispersion for Steady Flow in a Channel

For steady flow between two flat plates, the corresponding species conservation equation with an observer placed on the mean speed of the flow, i.e. $2/3 u_0$ is

$$\frac{\partial^2 c}{\partial y^2} = \frac{4h^2 u_0}{D} \left(y - y^2 - \frac{1}{6} \right) \frac{\partial \bar{c}}{\partial x} \tag{2.57}$$

The transverse coordinate, y , is normalized with the gap width, h , and the pseudo-steady approximation made by Taylor along with the negligible axial diffusion approximation are used in Equation 2.57. The solution to c given that the walls of the plates are impermeable to the diffusing species is then

$$c = \frac{\frac{2}{3}h^2 u_0}{D} \left(y^3 - \frac{1}{2}y^4 - \frac{1}{2}y^2 \right) \frac{\partial c_m}{\partial x_1} + k \quad (2.58)$$

where k as an unknown constant that cannot be determined using the given boundary conditions, yet it is not needed in order to find the effective diffusion coefficient. To find the mass transfer due to convection, the product of the average velocity and concentration is spatially integrated over the gap width. From this, the effective diffusion coefficient for flow between two flat plates is found to be

$$D_{eff} = \frac{2u_0^2 h^2}{945D} \quad (2.59)$$

2.4.4 Moment Justification in Two-Dimensional Geometry

To solve the above case by the method of moments, eigenfunction expansions will be used. The species conservation equation in a rectangular geometry is

$$\frac{\partial c}{\partial t} + V_x \frac{\partial c}{\partial x} = D \frac{\partial^2 c}{\partial y^2} + D \frac{\partial^2 c}{\partial x^2} \quad (2.60)$$

and the effective species conservation equation is unchanged from Equation 2.51 for the circular geometry. This means that the first three moment equation for the mean concentration are exactly Equations 2.53, 2.54, and 2.55. The corresponding moment equations for the concentration given in Equation 2.60 in scaled form are

$$\begin{aligned} \frac{\partial c_0}{\partial t} &= \frac{\partial^2 c_0}{\partial y^2} \\ \frac{\partial c_1}{\partial t} &= V_x c_0 + \frac{\partial^2 c_1}{\partial y^2} \\ \frac{\partial c_2}{\partial t} &= 2V_x c_1 + 2c_0 + \frac{\partial^2 c_2}{\partial y^2} \end{aligned} \quad (2.61, 2.62, 2.63)$$

where y is again normalized with h , t is scaled with h^2/D , V_x by D/h , and an effective diffusion coefficient scaled by the molecular diffusivity, D .

Eigenfunction expansions require the use of an inner product defined for this problem as

$$\langle a, b \rangle = \int_0^1 a \tilde{b} dy \quad (2.64)$$

where $\tilde{b}$ is the complex conjugate of b . In this problem, it turns out that $b = \tilde{b}$ and our inner product can also be seen as the mean, or spatial average, of a and b .

$$\langle a, b \rangle = \overline{ab} \quad (2.65)$$

The corresponding eigenvalue equations are

$$\begin{aligned} A\varphi_j &= \lambda_j^2 \varphi_j \\ A^* \psi_j &= \tilde{\lambda}_j^2 \psi_j \end{aligned} \quad (2.66, 2.67)$$

where A is the linear operator of our equation and A^* is the adjoint of A . The functions φ and ψ are orthogonal such that

$$\langle \varphi_j, \psi_k \rangle = \delta_{jk} \quad (2.68)$$

where

$$\delta_{kk} = \delta_{jj} = 1 \quad (2.69)$$

otherwise

$$\delta_{jk} = 0 \quad (2.70)$$

It can be proven that

$$c_m = \sum_{j=0}^{\infty} \langle \psi_j, c_m \rangle \varphi_j \quad (2.71)$$

For this problem, $A = \frac{\partial^2}{\partial y^2}$ which can be shown to be the same as its adjoint, A^* , with the

homogenous boundary conditions of the problem

$$\frac{\partial c_m}{\partial y} = \frac{\partial \varphi_j}{\partial y} = 0 \text{ at } y = 0 \text{ and } y = 1. \quad (2.72, 2.73)$$

Because $A = A^*$ and, as it happens, the eigenvalues are real, then $\varphi_j = \psi_j$. It can be easily shown that

$$\varphi_j = \sqrt{2} \cos(j\pi y) \text{ for } j \neq 0 \quad (2.74)$$

and

$$\varphi_0 = 1 \quad (2.75)$$

A problem of the form

$$\frac{\partial c_m}{\partial t} = \frac{\partial^2 c_m}{\partial y^2} + f(y, t) \quad (2.76)$$

can be solved using the corresponding eigenvalue equation, inner products, and an integrating factor to produce

$$\langle \varphi_j, c_m \rangle \exp\{\lambda_j^2 t\} = \int_0^t \langle \varphi_j, f(y, \tau) \rangle \exp\{\lambda_j^2 \tau\} d\tau + \text{constant} \quad (2.77)$$

where $\lambda_j = j\pi$.

Solving for c_0 in Equation 2.61 with $f(y, t) = 0$ gives

$$c_0 = \sum_{j=0}^{\infty} \langle \varphi_j, c_0(t=0) \rangle \varphi_j \exp\{-\lambda_j^2 t\} \quad (2.78)$$

where $c_0(t=0)$ is the value of c_0 at time zero. Solving for c_1 with $f(y, t) = V_x c_0$ gives

$$\begin{aligned} c_1 = & \sum_{k=0}^{\infty} \sum_{j=0}^{\infty} \langle \varphi_k, V_x \varphi_j \rangle \langle \varphi_j, c_0(t=0) \rangle \frac{\exp\{-\lambda_j^2 t\} - \exp\{-\lambda_k^2 t\}}{\lambda_k^2 - \lambda_j^2} \varphi_k \\ & + \sum_{k=0}^{\infty} \langle \varphi_k, c_1(t=0) \rangle \varphi_k \exp\{-\lambda_k^2 t\} \end{aligned} \quad (2.79)$$

where $\lambda_k = k\pi$ and $c_1(t=0)$ is the initial value of c_1 . Next, the concentration must be spatially averaged in order to find its relation to the mean concentration equations in order to evaluate D_{eff} and V_{eff} .

In finding the mean concentration of the moments, the average of the above moment equations must be found using

$$\begin{aligned}\overline{\frac{\partial c_0}{\partial t}} &= \overline{\frac{\partial^2 c_0}{\partial y^2}} \\ \overline{\frac{\partial c_1}{\partial t}} &= \overline{V_x c_0} + \overline{\frac{\partial^2 c_1}{\partial y^2}} \\ \overline{\frac{\partial c_2}{\partial t}} &= 2\overline{V_x c_1} + \overline{2c_0} + \overline{\frac{\partial^2 c_2}{\partial y^2}}\end{aligned}\tag{2.80, 2.81, 2.82}$$

It can be shown by using integration by parts and the appropriate boundary conditions that

$$\overline{\frac{\partial^2 c_m}{\partial y^2}} = 0\tag{2.83}$$

so that the equations become

$$\begin{aligned}\overline{\frac{\partial \bar{c}_0}{\partial t}} &= 0 \\ \overline{\frac{\partial \bar{c}_1}{\partial t}} &= \overline{V_x c_0} \\ \overline{\frac{\partial \bar{c}_2}{\partial t}} &= 2\overline{V_x c_1} + 2\bar{c}_0\end{aligned}\tag{2.84, 2.85, 2.86}$$

therefore, the equations for V_{eff} and D_{eff} become

$$\begin{aligned}V_{eff} &= \frac{\overline{V_x c_0}}{\bar{c}_0} \\ D_{eff} &= 1 + \frac{\overline{V_x c_1}}{\bar{c}_0} - \frac{V_{eff} \bar{c}_1}{\bar{c}_0}\end{aligned}\tag{2.87, 2.88}$$

To find V_{eff} , it is first noted that

$$\bar{c}_0 = \bar{c}_0(t=0) \quad (2.89)$$

and

$$\overline{V_x c_0} = \int_0^1 V_x c_0 dy = \sum_{j=0}^{\infty} \langle \varphi_j, c_0(t=0) \rangle \overline{V_x \varphi_j} \exp[-\lambda_j^2 t] = \bar{c}_0(t=0) \bar{V}_x + O(\exp[-\lambda_1^2 t]) \quad (2.90)$$

For long times after the initial transients die out, the higher order terms become small as t becomes large and the exponentials approach zero. Thus, the effective velocity becomes

$$V_{eff} = \bar{V}_x \quad (2.91)$$

To find D_{eff} , two mean values are needed and are of the form

$$\bar{c}_1 = \bar{c}_1(t=0) + \bar{V}_x \bar{c}_0(t=0) + \sum_{j=0}^{\infty} \overline{V_x \varphi_j} \langle \varphi_j, c_0(t=0) \rangle \frac{1}{\lambda_j^2} + O(\exp[-\lambda_1^2 t]) \quad (2.92)$$

and

$$\begin{aligned} \overline{V_x c_1} &= \sum_{k=0}^{\infty} \sum_{j=0}^{\infty} \langle \varphi_k, V_x \varphi_j \rangle \langle \varphi_j, c_0(t=0) \rangle \frac{\exp[-\lambda_j^2 t] - \exp[-\lambda_k^2 t]}{\lambda_k^2 - \lambda_j^2} \overline{V_x \varphi_k} + \sum_{k=0}^{\infty} \langle \varphi_k, c_1(t=0) \rangle \overline{\varphi_k V_x} \exp[-\lambda_k^2 t] \\ &= \bar{V}_x \bar{c}_0(t=0) \bar{V}_x + \sum_{k=0}^{\infty} \langle \varphi_k, V_x \rangle \bar{c}_0(t=0) \frac{1 - \exp[-\lambda_k^2 t]}{\lambda_k^2} \overline{\varphi_k V_x} + \sum_{j=0}^{\infty} \overline{V_x \varphi_j} \langle \varphi_j, c_0(t=0) \rangle \bar{V}_x \frac{\exp[-\lambda_j^2 t] - 1}{\lambda_j^2} \\ &\quad + \bar{c}_1(t=0) \bar{V}_x + O(\exp[-\lambda_1^2 t]) + O(t \exp[-\lambda_1^2 t]) \end{aligned} \quad (2.93)$$

For long times, the exponential terms are essentially zero, and the effective diffusion coefficient becomes

$$D_{eff} = 1 + \sum_{k=1}^{\infty} \frac{\langle V_x, \varphi_k \rangle \langle V_x, \varphi_k \rangle}{\lambda_k^2} \quad (2.94)$$

To evaluate $\int_0^1 \varphi_k V_x dy$, first see that

$$\int_0^1 \varphi_k \frac{d^2 V_x}{dy^2} dy = \left[\varphi_k \frac{dV_x}{dy} \right]_0^1 - \left[\frac{d\varphi_k}{dy} \frac{dV_x}{dy} \right]_0^1 + \int_0^1 \frac{d^2 \varphi_k}{dy^2} V_x dy \quad (2.95)$$

With $V_x = 4u_0(y - y^2)$ as the normalized velocity and using Equation 2.74 for ϕ_k , this gives

$$0 = \left[4\sqrt{2}u_0(1 - 2y)\cos(k\pi y) \right]_0^1 - \lambda_k^2 \int_0^1 \phi_k V_x dy \quad (2.96)$$

from which

$$\int_0^1 \phi_k V_x dy = \langle V_x, \phi_k \rangle = -\frac{4\sqrt{2}u_0}{\lambda_k^2} [(-1)^k + 1] \quad (2.97)$$

Using this in Equation 2.92 for D_{eff} and recalling that $\lambda_k = k\pi$, D_{eff} then becomes

$$D_{eff} = 1 + \frac{64u_0^2}{\pi^6} \sum_{k=1}^{\infty} \left[\frac{1 + (-1)^k}{k^6} \right] \quad (2.98)$$

Evaluating the summation series and reverting back to unscaled quantities, the final expression for D_{eff} is

$$D_{eff} = D + \frac{2h^2u_0^2}{945D} \quad (2.99)$$

which is exactly the expression derived using Taylor's method save the additional term D which now accounts for pure molecular diffusion.

2.4.5 Solution for Periodic Case in a Channel Using Eigenfunction Expansions

Now that Taylor's method and the method of moments have been proven to correctly predict the effective diffusion coefficient for a rectangular geometry, moments will be employed to justify the concentration approximation used in calculating the concentration field in periodic flow between two flat plates. First, however, the solution to the periodic flow problem will be re-derived using eigenfunction expansions. This form of the solution will easily relate to the solution derived using the method of moments that utilize eigenfunction expansions in their solution. The species conservation equation for a rectangular geometry given by Equation 2.60 will again be used with the

appropriate approximations and boundary conditions. The same definition for the inner product will be used as in the steady flow case, and the corresponding eigenvalue equations will also be the same with φ_k and φ_0 defined in Equation 2.74 and Equation 2.75 respectively. Using Equation 2.77 as a guide to find the inner product and defining the velocity as

$$V_x = \hat{V}(y)e^{i\omega t} + \tilde{V}(y)e^{-i\omega t} \quad (2.100)$$

the inner products become

$$\langle \varphi_k, c^* \rangle = -\frac{\Delta c}{L} \left[\langle \varphi_k, \hat{V} \rangle \frac{e^{i\omega t} - e^{-\lambda_k^2 t}}{\lambda_k^2 + i\omega} + \langle \varphi_k, \tilde{V} \rangle \frac{e^{-i\omega t} - e^{-\lambda_k^2 t}}{\lambda_k^2 - i\omega} \right] + \langle \varphi_k, c^*(t=0) \rangle e^{-\lambda_k^2 t} \quad (2.101)$$

and

$$\langle \varphi_0, c^* \rangle = -\frac{\Delta c}{L} \left[\overline{\tilde{V}} \frac{e^{i\omega t} - 1}{i\omega} + \overline{\tilde{V}} \frac{e^{-i\omega t} - 1}{-i\omega} \right] + \overline{c^*(t=0)} \quad (2.102)$$

where the overbar again denotes the transverse spatial average. After long times, the expression for the concentration is

$$\begin{aligned} c = & \frac{\Delta c}{L} x + \overline{c^*(t=0)} - \frac{\Delta c}{L} \left[\overline{\tilde{V}} \frac{e^{i\omega t} - 1}{i\omega} + \overline{\tilde{V}} \frac{e^{-i\omega t} - 1}{-i\omega} \right] \\ & - \frac{\Delta c}{L} \sum_{k=1}^{\infty} \left[\langle \varphi_k, \hat{V} \rangle \frac{e^{i\omega t}}{\lambda_k^2 + i\omega} + \langle \varphi_k, \tilde{V} \rangle \frac{e^{-i\omega t}}{\lambda_k^2 - i\omega} \right] \varphi_k \end{aligned} \quad (2.103)$$

The time averaged convective axial flux will be calculated as before by time averaging the product of the velocity and concentration to give

$$J_{conv}^{avg} = -\frac{\Delta c}{L} \left[\overline{\tilde{V}\hat{V}} - \overline{\hat{V}\tilde{V}} \right] - \frac{\Delta c}{L} \sum_{k=1}^{\infty} \left[\langle \varphi_k, \hat{V} \rangle \frac{\tilde{V}}{\lambda_k^2 + i\omega} + \langle \varphi_k, \tilde{V} \rangle \frac{\hat{V}}{\lambda_k^2 - i\omega} \right] \varphi_k \quad (2.104)$$

Finally, the flux is spatially averaged to give the final expression for the convective mass transfer

$$Q_{conv}^{avg} = -\frac{\Delta c}{L} \sum_{k=1}^{\infty} \left[\frac{\langle \varphi_k, \tilde{V} \rangle \langle \varphi_k, \hat{V} \rangle}{\lambda_k^2 + i\omega} + \frac{\langle \varphi_k, \tilde{V} \rangle \langle \varphi_k, \hat{V} \rangle}{\lambda_k^2 - i\omega} \right] \quad (2.105)$$

2.4.6 Moment Verification for Periodic Flow in a Channel

Now that the mass transfer has been found using eigenfunction expansions and the appropriate approximations, it will be determined for oscillating plates this time using power moments. Again, the equations that need to be solved are Equations 2.61, 2.62, and 2.63, and D_{eff} is still defined in Equation 2.94. The primary difference between this calculation and previous calculations is found in determining c_1 by using the proper integrating factor used in Equation 2.97. This time, the velocity is not constant, but is a periodic function of time defined in Equation 2.100 so that

$$\begin{aligned} \langle \varphi_j, c_1 \rangle &= e^{-\lambda_j^2 t} \int_0^t \left\langle \varphi_j, \hat{V} c_0(\tau) \right\rangle e^{(i\omega + \lambda_j^2)\tau} + \left\langle \varphi_j, \tilde{V} c_0(\tau) \right\rangle e^{(-i\omega + \lambda_j^2)\tau} \Big| d\tau \\ &+ \langle \varphi_j, V_x c_0(t=0) \rangle e^{-\lambda_j^2 t} \end{aligned} \quad (2.106)$$

Using c_0 given in Equation 2.78,

$$\langle \varphi_j, c_1 \rangle = \langle \varphi_j, V_x c_0(t=0) \rangle e^{-\lambda_j^2 t} + \sum_{k=0}^{\infty} \left[\left\langle \varphi_j, \hat{V} \varphi_k \right\rangle \langle \varphi_k, c_0(t=0) \rangle \frac{e^{(i\omega - \lambda_k^2)t} - e^{-\lambda_j^2 t}}{i\omega + \lambda_j^2 - \lambda_k^2} + \left\langle \varphi_j, \tilde{V} \varphi_k \right\rangle \langle \varphi_k, c_0(t=0) \rangle \frac{e^{(-i\omega - \lambda_k^2)t} - e^{-\lambda_j^2 t}}{-i\omega + \lambda_j^2 - \lambda_k^2} \right] \quad (2.107)$$

Using the definition of c_1 given in Equation 2.92 with $m = 1$, c_1 becomes

$$\begin{aligned} c_1 &= \sum_{j=0}^{\infty} \langle \varphi_j, V_x c_0(t=0) \rangle e^{-\lambda_j^2 t} \varphi_j + \sum_{j=0}^{\infty} \sum_{k=0}^{\infty} \left[\left\langle \varphi_j, \hat{V} \varphi_k \right\rangle \langle \varphi_k, c_0(t=0) \rangle \frac{e^{(i\omega - \lambda_k^2)t} - e^{-\lambda_j^2 t}}{i\omega + \lambda_j^2 - \lambda_k^2} \varphi_j \right. \\ &\quad \left. + \left\langle \varphi_j, \tilde{V} \varphi_k \right\rangle \langle \varphi_k, c_0(t=0) \rangle \frac{e^{(-i\omega - \lambda_k^2)t} - e^{-\lambda_j^2 t}}{-i\omega + \lambda_j^2 - \lambda_k^2} \varphi_j \right] \end{aligned} \quad (2.108)$$

Observe that $\bar{c}_1$ is given by

$$\bar{c}_1 = \langle \varphi_0, c_1 \rangle \quad (2.109)$$

since $\varphi_0 = 1$. Using this and the orthogonality of the eigenfunctions, that is

$$\varphi_0 \varphi_j = 0 \text{ if } j \neq 0 \quad (2.110)$$

and

$$\varphi_0 \varphi_j = 1 \text{ if } j = 0, \quad (2.111)$$

$\bar{c}_1$ becomes

$$\begin{aligned} \bar{c}_1 = & \bar{c}_1(t=0) + \bar{V} \bar{c}_0(t=0) \frac{e^{i\omega t} - 1}{i\omega} + \bar{V} \bar{c}_0(t=0) \frac{e^{-i\omega t} - 1}{-i\omega} \\ & + \sum_{k=1}^{\infty} \left[\overline{\hat{V} \varphi_k} \langle \varphi_k, c_0(t=0) \rangle \frac{e^{(i\omega - \lambda_k^2)t} - 1}{i\omega - \lambda_k^2} + \overline{\tilde{V} \varphi_k} \langle \varphi_k, c_0(t=0) \rangle \frac{e^{(-i\omega - \lambda_k^2)t} - 1}{-i\omega - \lambda_k^2} \right] \end{aligned} \quad (2.112)$$

Recalling that $\bar{c}_0 = \overline{c_0(t=0)}$, which is a constant, this makes

$$\begin{aligned} \overline{V_x c_0 c_1} = & \bar{V}_x \bar{c}_0(t=0) \bar{c}_1(t=0) + \bar{V}_x \bar{V} \bar{c}_0(t=0) \bar{c}_0(t=0) \frac{e^{i\omega t} - 1}{i\omega} + \bar{V}_x \bar{V} \bar{c}_0(t=0) \bar{c}_0(t=0) \frac{e^{-i\omega t} - 1}{-i\omega} \\ & + \bar{V}_x \bar{c}_0(t=0) \sum_{k=1}^{\infty} \left[\langle \varphi_k, \hat{V} \rangle \langle \varphi_k, c_0(t=0) \rangle \frac{e^{(i\omega - \lambda_k^2)t} - 1}{i\omega - \lambda_k^2} + \langle \varphi_k, \tilde{V} \rangle \langle \varphi_k, c_0(t=0) \rangle \frac{e^{(-i\omega - \lambda_k^2)t} - 1}{-i\omega - \lambda_k^2} \right] \end{aligned} \quad (2.113)$$

after initial transients die out, or as $t \rightarrow \infty$. In the same manner, after long times, the

expression for $\overline{V_x c_1}$ becomes

$$\begin{aligned} \overline{V_x c_1} = & \bar{c}_1(t=0) \bar{V}_x + \bar{V}_x \bar{V} \bar{c}_0(t=0) \frac{e^{i\omega t} - 1}{i\omega} + \bar{V}_x \bar{V} \bar{c}_0(t=0) \frac{e^{-i\omega t} - 1}{-i\omega} \\ & + \bar{V}_x \sum_{k=1}^{\infty} \left[\overline{\varphi_k} \hat{V} \langle \varphi_k, c_0(t=0) \rangle \frac{e^{(i\omega - \lambda_k^2)t} - 1}{i\omega - \lambda_k^2} + \overline{\varphi_k} \tilde{V} \langle \varphi_k, c_0(t=0) \rangle \frac{e^{(-i\omega - \lambda_k^2)t} - 1}{-i\omega - \lambda_k^2} \right] \\ & + \sum_{j=1}^{\infty} \left[\overline{\varphi_j V_x} \langle \varphi_j, \hat{V} \rangle \bar{c}_0(t=0) \frac{e^{i\omega t} - 1}{i\omega + \lambda_j^2} + \overline{\varphi_j V_x} \langle \varphi_j, \tilde{V} \rangle \bar{c}_0(t=0) \frac{e^{-i\omega t} - 1}{-i\omega + \lambda_j^2} \right] \end{aligned} \quad (2.114)$$

Because $\overline{ab} = \langle a, b \rangle$, Equation 2.94 for D_{eff} is then

$$D_{eff} = 1 + \frac{\overline{V_x c_1}}{\bar{c}_0} - \frac{\overline{V_x c_0 \bar{c}_1}}{\bar{c}_0 \bar{c}_0} = 1 + \sum_{j=1}^{\infty} \left[\langle V_x, \varphi_j \rangle \langle \varphi_j, \hat{V} \rangle \frac{e^{i\omega} - 1}{i\omega + \lambda_j^2} + \langle V_x, \varphi_j \rangle \langle \varphi_j, \tilde{V} \rangle \frac{e^{-i\omega} - 1}{-i\omega + \lambda_j^2} \right] \quad (2.115)$$

Recalling that $V_x = \hat{V}e^{i\omega t} + \tilde{V}e^{-i\omega t}$, taking the time average of the above expression will give the final expression for the effective diffusion coefficient.

$$D_{eff}^{avg} = 1 + \sum_{j=1}^{\infty} \left[\frac{\langle \tilde{V}, \varphi_j \rangle \langle \varphi_j, \hat{V} \rangle}{i\omega + \lambda_j^2} + \frac{\langle \hat{V}, \varphi_j \rangle \langle \varphi_j, \tilde{V} \rangle}{-i\omega + \lambda_j^2} \right] \quad (2.116)$$

Knowing that $Q^{avg} = -\frac{\Delta c}{L} D_{eff}^{avg}$ for the flat plates, it can be seen that the convective mass transfer derived using the power moments is precisely the same as the convective mass transfer found in Equation 2.105 using the concentration approximation. This shows that the concentration approximation is indeed valid after long times and is an appropriate assumption in periodic flow problems.

CHAPTER 3

PULSATILE FLOW IN CIRCULAR GEOMETRIES-OPEN TUBES AND ANNULAR REGIONS

The phenomena observed in the two-dimensional case of periodic flow in a channel will also be true for a three-dimensional, circular geometry. The quantitative values may be different, but the qualitative results and the explanation of the physics still hold for circular geometries. This chapter will focus on periodic flow in two specific circular configurations: flow in an open tube and flow in an annular region between two tubes. Harris and Goren [3] studied periodic flow in an open tube and Watson [4] gave some general results for periodic flow in various geometries with a result given for the special case of an open tube. Kurzweg, Jaeger, and collaborators [6-11] performed much of their research with tubes or bundles of tubes of small diameters. Beal [21] and Rader [22] studied more complex circular geometries and much of Poplasky's [1] work focused on annular flow. The analysis presented in this chapter will be periodic flow driven by an oscillating boundary. With the exception of Rader [22] and Poplasky [1], the periodic flow of the authors mentioned has primarily been induced by an oscillating pressure drop. As already shown in the previous chapter, both methods are similar in nature. This chapter will seek to compare the mass transfer and separation one can achieve in both circular configurations along with further analysis of imperfections associated with periodic flow in an annular region. In particular, explanations will be given on the effect of a distorted annular region on the mass transfer and separation.

3.1 Evaluation of the Total Mass Transfer

The mass transfer is evaluated in the same way as already outlined in periodic flow in a channel. Because both the open tube and annular geometry have been studied previously, only the governing equations and final result for the total mass transfer will be presented in this section.

A depiction of both circular geometries is given in Fig. 3.1. For this analysis, it will be assumed that the cross-sectional area of both configurations are the same. The equation of motion for a boundary driven configuration in cylindrical coordinates is now

$$\frac{\partial V_z}{\partial t} = \frac{v}{r} \frac{\partial}{\partial r} \left(r \frac{\partial V_z}{\partial r} \right) \quad (3.1)$$

For an open tube, the no slip boundary conditions are

$$V_z = \frac{1}{2} A \omega \cos(\omega t) \text{ at } r = R \quad (3.2)$$

and

$$V_z \text{ is finite at } r = 0 \quad (3.3)$$

For an annular geometry, the boundary conditions are

$$V_z = \frac{1}{2} A \omega \cos(\omega t) \text{ at } r = R_{out} \quad (3.4)$$

and

$$V_z = \frac{1}{2} A \omega \cos(\omega t) \text{ at } r = R_{in} \quad (3.5)$$

The species conservation equation is

$$\frac{\partial c}{\partial t} + V_z \frac{\partial c}{\partial z} = D \left[\frac{\partial^2 c}{\partial r^2} + \frac{1}{r} \frac{\partial c}{\partial r} + \frac{\partial^2 c}{\partial z^2} \right] \quad (3.6)$$

with the boundaries assumed to be impermeable to the diffusing species with the added condition for the open tube that the concentration is finite at $r = 0$. The concentration can

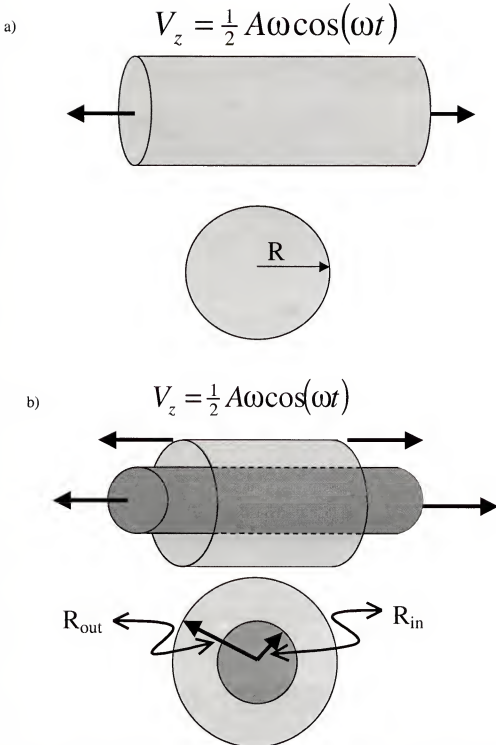


Figure 3.1 a) Open tube geometry of radius R . b) Annular geometry with outer radius R_{out} and inner radius R_{in} .

be again written as the sum of an axial term plus a term depending only on the radial position and time. This was proven to be an appropriate assumption in the previous chapter using the method of moments and can be presumed to be true here as well.

Finally, the total time-averaged mass transfer for an open tube is

$$\bar{Q} = -\frac{D(c_2 - c_1)\pi R^2}{L} + \frac{\omega}{2\pi} \int_0^{\frac{2\pi}{\omega}} \int_0^{2\pi} \int_0^R V_z C r dr d\theta dt \quad (3.7)$$

and for an annulus

$$\bar{Q} = -\frac{D(c_2 - c_1)}{L} \pi (R_{out}^2 - R_{in}^2) + \frac{\omega}{2\pi} \int_0^{\frac{2\pi}{\omega}} \int_0^{2\pi} \int_{R_{in}}^{R_{out}} V_z C r dr d\theta dt \quad (3.8)$$

The full expression for the total mass transfer of the open tube geometry is given in Appendix B. As a result of the complicated and unwieldy nature of the expression for total mass transfer of the annular configuration, nothing is gained upon its observation.

Only graphs constructed by Maple® are needed for comparison between the two geometries. If one is still interested in the calculated result, I will refer the reader to Equation 102 of Poplasky's thesis [1] or Equation 86 of Rader's report [22] for the full expression of the total mass transfer for an annulus with both tubes oscillating in phase.

3.2 Comparing the Total Mass Transfer and Separation for Both Circular Configurations

In order to properly compare the open tube and annular configurations, the cross sectional areas of both are taken to be the same. The geometrical parameters of the system for the following results are given in Table 3.1. The parameters and fluid properties not explicitly given in Table 3.1 can be found in Table 2.1. The values for the inner and outer radius for the annular geometry were chosen because of the limitations associated with Maple® in evaluating the complex groupings of Bessel functions.

Maple® is unable to evaluate large values in the argument of the Bessel functions in

these complex expressions and trying to replace the functions with asymptotic expansions has proven difficult. In this case, it was much easier to choose parameters that gave appropriate results for a reasonable range of frequencies.

Table 3.1 System properties used in example calculations

Symbol	Name	Value	Units
A	Peak-to-peak amplitude for both configuration	10	cm
R	Open tube radius	0.995	cm
R_{out}	Annular outer radius	1.0	cm
R_{in}	Annular inner radius	0.1	cm
$(c_1 - c_2) / L$	Normalized concentration difference with plate length ¹	1.0	mol/cm ⁴

The mass transfer for the open tube geometry shown in Fig. 3.2 for a helium and CO₂ binary system in a nitrogen carrier reveals that multiple crossover frequencies are still attainable as the case for periodic flow in a channel. The ratio of the slow diffuser to the fast diffuser depicted in Fig. 3.3 also shows the same trends seen previously in the two-dimensional geometry. For an annular geometry, Fig. 3.4 shows the total mass transfer for the same system with a single crossover for the range of frequencies presented. If the software was able to plot the higher frequencies, it is reasonable to assume that a second crossover would also be evident. The measure of the separation given in Fig. 3.5 also shows similar results. This indicates that the physical arguments presented in the previous chapter also apply to these geometries.

In comparing the two geometrical configurations, it is evident in Fig. 3.6 that the total time-averaged mass transfer for the open tube is greater than that for the annulus

¹ This normalized value is used for calculation purposes only. In reality, this concentration difference would violate the assumption that the species are dilute. In this section, it is only the qualitative results that are of interest.

over all frequencies (except when $\omega = 0$ when they are the same). This demonstrates that for greater throughput for a single species, an open tube geometry is more efficient. For

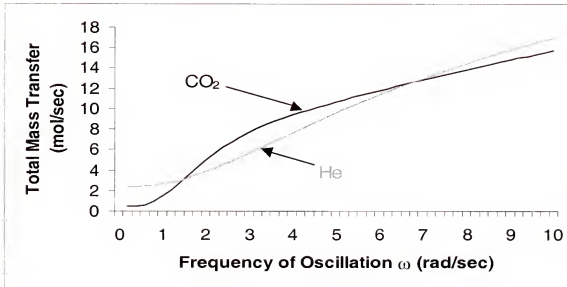


Figure 3.2 Total mass transfer versus frequency for carbon dioxide and helium in an open tube geometry.

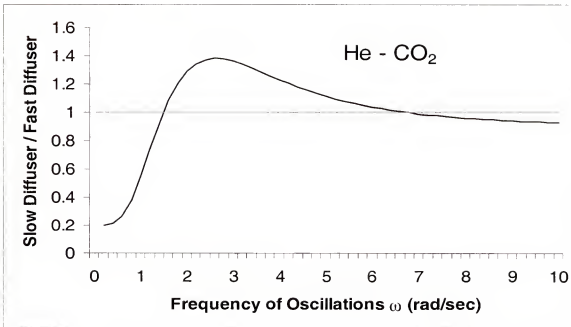


Figure 3.3 Separation ratio for helium and carbon dioxide in the open tube configuration.

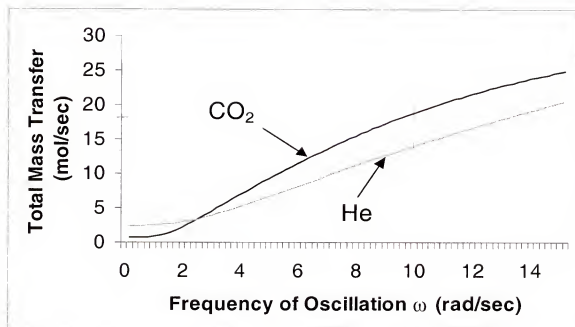


Figure 3.4 Total mass transfer versus frequency for carbon dioxide and helium in an annular configuration.

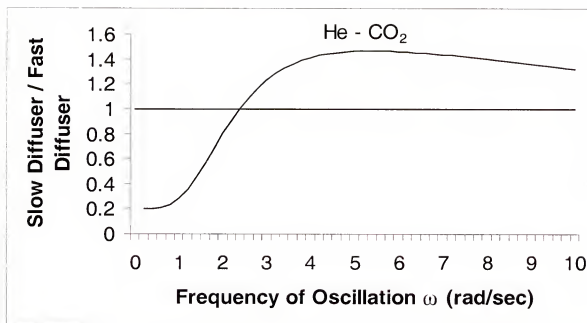


Figure 3.5 Separation ratio for carbon dioxide and helium for the annular configuration.

the purposes of explaining the physics why this is true, imagine that an oscillating *pressure drop* is creating the periodic flow instead of the oscillating boundaries. This would make regions of slow flow near the boundaries of each system. For an open tube, there is only one boundary near where the fluid velocity is small and the region of fast moving fluid is in the core of the tube. For an annulus, there is also a slow flow region along the outer tube like the open tube configuration, but there is an added slow flow region near the boundary of the inner tube. This extra slow flow region will hinder the axial transport of species in an annulus more than what would occur in an open tube.

Thus, the mass transfer will be greater for an open tube geometry.

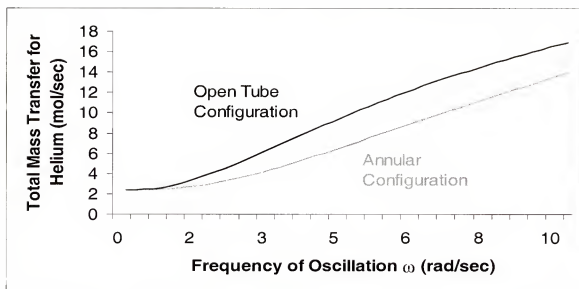


Figure 3.6 Total mass transfer for helium in an open tube and annular configuration.

The separation achieved for both geometries for a He-CO₂ binary system and an He-CH₄ binary system are shown in Fig. 3.7 and Fig. 3.8 respectively. For the annular geometry, the maximum in the separation of the slow diffusing species (CO₂ or CH₄) is slightly higher than the maximum in the open tube. Further, as the maximum occurs at a

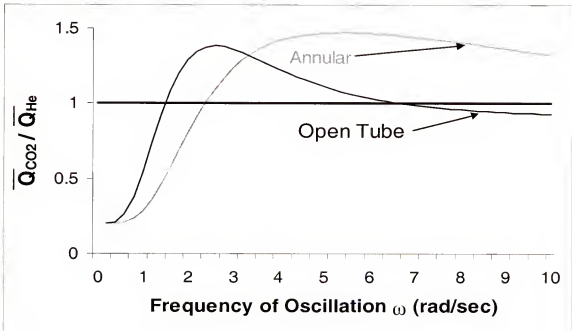


Figure 3.7 Separation ratios for a carbon dioxide and helium system for both circular configurations.

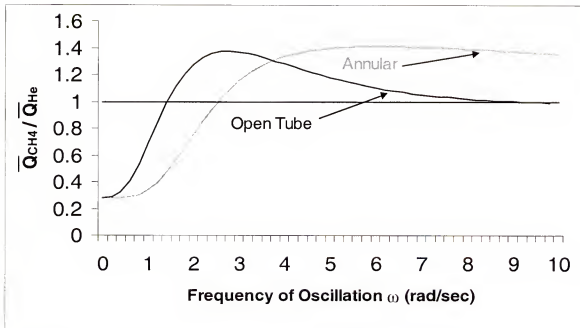


Figure 3.8 Separation ratio for a methane and helium system for both circular configurations.

higher frequency for the annulus, the mass transfer of both dilute species is higher for the annular configuration at this point as well. This indicates that an annular configuration will give a greater maximum separation with a higher throughput of species than the open tube geometry even though the mass transfer for each species in the open tube geometry is higher for all frequencies. This motivates the use of an annular geometry as a viable method to achieve a reasonable separation at a high throughput.

3.3 The Effect of Eccentricity on Periodic Annular Flow

Now that the relationship between periodic flow in an open tube and in an annular configuration has been modeled, experiments will also be conducted for both configurations. In addition to the above analysis, Beal [21], Poplasky [1], and Rader [22], have all done theoretical studies on periodic annular flow. Poplasky [1] has also performed experiments to verify his predictions and found reasonable agreement in his study. However, during experiments, it is difficult to ensure that the inner tube is perfectly centered within the outer tube. Therefore, before experimental results are presented, an analysis on the effect of eccentricity, or the inner tube being slightly off center, on the mass transfer and separation produced by periodic annular flow is studied. Regular perturbation techniques will be employed to find the new velocity and concentration, thus giving rise to the mass transfer in the new eccentric geometry.

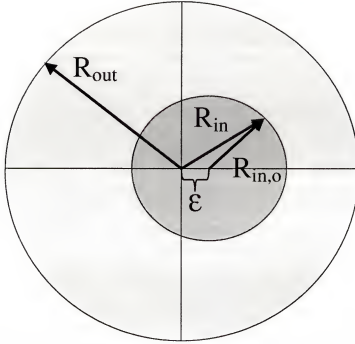


Figure 3.9 Eccentric annular geometry with inner cylinder displaced from the center of the outer cylinder by an amount ϵ . R_{in} is now the new inner radius of the displaced inner cylinder.

Now, suppose that in the annular configuration the inner tube is slightly displaced from the center of the outer tube by an amount ϵ as shown in Fig. 3.9. The outer radius remains unchanged, but the inner radius is now a function of ϵ by way of the Taylor series expansion about $\epsilon = 0$.

$$R_{in}(\epsilon) = R_0 + \epsilon R_1 + \frac{\epsilon^2}{2!} R_2 + \dots \quad (3.9)$$

In Fig. 3.9, the law of cosines dictates that

$$R_{in,0}^2 = R_{in}^2 + \epsilon^2 - 2\epsilon R_{in} \cos \theta \quad (3.10)$$

where $R_{in,0}$ is the radius of the inner cylinder in the unperturbed state. Substituting the definition of $R_{in}(\epsilon)$ into the above equation gives

$$R_{in,0}^2 = \left(R_0^2 + \epsilon^2 R_1^2 + 2\epsilon R_0 R_1 + \epsilon^2 R_0 R_2 \right) + \epsilon^2 - 2\epsilon (R_0 + \epsilon R_1) \cos \theta + O(\epsilon^3) \quad (3.11)$$

Equating like orders of ε gives the following relations

$$R_0 = R_{m,0} \quad (3.12)$$

$$R_1 = \cos \theta \quad (3.13)$$

$$R_2 = \frac{\cos^2 \theta - 1}{R_0} \quad (3.14)$$

As the boundary has shifted slightly from its centered base state, the radial position variable, r , is redefined as a function of ε with the Taylor series expansion

$$r = r_0 + \varepsilon r_1 + \frac{\varepsilon^2}{2!} r_2 + \dots \quad (3.15)$$

In a sense, the domain region between the two tubes and the inner boundary have been “remapped” because of the shift. The other position variables in cylindrical coordinates and the time variable, t , remain unchanged in the new geometry as

$$z = z_0, \quad (3.16)$$

$$\theta = \theta_0, \quad (3.17)$$

and

$$t = t_0 \quad (3.18)$$

3.3.1 Perturbed Velocity Equations

The z -component of the velocity can now be written as a Taylor series expansion about $\varepsilon = 0$ as

$$V_z(r, \theta, t, \varepsilon) = V_0 + \varepsilon \left. \frac{dV_z}{d\varepsilon} \right|_{\varepsilon=0} + \frac{\varepsilon^2}{2!} \left. \frac{d^2 V_z}{d\varepsilon^2} \right|_{\varepsilon=0} + \dots \quad (3.19)$$

However, not only is the velocity directly a function of ε as immediately seen in the series expansion, it is also implicitly a function of ε through the position variable r as

given in Equation 3.15. Because total derivatives of the velocity with respect to ε are needed, the “mapping” of the new geometry needs to be taken into account. Johns and Narayanan [23] in their second essay give a detailed explanation on the mapping for systems whose boundaries have been altered from a given base state. According to their conclusions, the Taylor series expansion for the velocity will become

$$V_z(r, \theta, t, \varepsilon) = V_0 + \varepsilon \left(V_1 + r_1 \frac{\partial V_0}{\partial r_0} \right) + \frac{1}{2} \varepsilon^2 \left(V_2 + \frac{\partial^2 V_0}{\partial r_0^2} r_1^2 + 2 \frac{\partial V_1}{\partial r_0} + \frac{\partial V_0}{\partial r_0} r_2 \right) + \dots \quad (3.20)$$

where

$$V_1 = \left. \frac{\partial V_z}{\partial \varepsilon} \right|_{\varepsilon=0} \quad (3.21)$$

and

$$V_2 = \left. \frac{\partial^2 V_z}{\partial \varepsilon^2} \right|_{\varepsilon=0} \quad (3.22)$$

Order ε^0

The velocity equations for the zeroth order represent the velocity for a centered inner tube. The domain equations are

$$\frac{\partial V_0}{\partial t_0} = \frac{v}{r_0} \frac{\partial}{\partial r_0} \left(r_0 \frac{\partial V_0}{\partial r_0} \right) \quad (3.23)$$

with the following boundary conditions

$$V_0 = \frac{1}{2} A \omega \cos(\omega t) \text{ at } r_0 = R_0 \quad (3.24)$$

$$V_0 = \frac{1}{2} A \omega \cos(\omega t) \text{ at } r_0 = R_{out} \quad (3.25)$$

As done previously, the velocity will be written as

$$V_0 = \hat{V}_0(r) e^{i\omega t_0} + \tilde{V}_0(r) e^{-i\omega t_0} \quad (3.26)$$

to give a solution

$$\hat{V}_0 = k_{01}J_0(i\lambda r_0) + k_{02}Y_0(i\lambda r_0) \quad (3.27)$$

and

$$\tilde{V}_0 = k_{03}J_0(\lambda r_0) + k_{04}Y_0(\lambda r_0) \quad (3.28)$$

where k_{01} , k_{02} , k_{03} , and k_{04} are constants. They can be determined from the boundary conditions and are given in the Appendix C. The value for λ is the same as given previously in Equation 1.7.

Order ϵ^1

The first order velocity perturbation equations are unaffected by the mapping in the domain except that the velocity is now a function of θ . However, the mapping does appear in the boundary equations because of the new geometry. As noted in the second essay of Johns and Narayanan [23], the boundary equations are referenced to the base, or unperturbed, state ($\epsilon = 0$). The domain equation is then

$$\frac{\partial V_1}{\partial r_0} = v \left(\frac{\partial^2 V_1}{\partial r_0^2} + \frac{1}{r_0} \frac{\partial V_1}{\partial r_0} + \frac{1}{r_0^2} \frac{\partial^2 V_1}{\partial \theta^2} \right) \quad (3.29)$$

with the corresponding boundary conditions

$$V_1 = 0 \text{ at } r_0 = R_{\text{out}} \quad (3.30)$$

$$V_1 + R_1 \frac{\partial V_0}{\partial r_0} = 0 \text{ at } r_0 = R_0 \quad (3.31)$$

Noting that $R_1 = \cos \theta$, V_1 must be of the form

$$V_1 = \hat{V}_1 e^{i\omega t_0} \cos \theta + \tilde{V}_1 e^{-i\omega t_0} \cos \theta \quad (3.32)$$

The solution to the first order velocity is then

$$\hat{V}_1 = k_{11}J_1(i\lambda r_0) + k_{12}Y_1(i\lambda r_0) \quad (3.33)$$

and

$$\tilde{V}_1 = k_{13} J_1(\lambda r_0) + k_{14} Y_1(\lambda r_0) \quad (3.34)$$

where the constants again are found from the boundary conditions and are given in Appendix C.

Order ϵ^2

As in the first order velocity equation, the mapping is seen only in the boundary equations and not in the domain equation for the second order velocity. The resulting equations for the second order velocity are for the domain

$$\frac{\partial V_2}{\partial t_0} = v \left(\frac{\partial^2 V_2}{\partial r_0^2} + \frac{1}{r_0} \frac{\partial V_2}{\partial r_0} + \frac{1}{r_0^2} \frac{\partial^2 V_2}{\partial \theta_0^2} \right) \quad (3.35)$$

and on the boundary

$$V_2 = 0 \quad \text{at } r_0 = R_{\text{out}} \quad (3.36)$$

$$V_2 + \frac{\partial^2 V_0}{\partial r_0^2} R_1^2 + 2 \frac{\partial V_1}{\partial r_0} R_1 + \frac{\partial V_0}{\partial r_0} R_2 = 0 \quad \text{at } r_0 = R_0 \quad (3.37)$$

Looking at the second boundary condition, it is noted that R_2 is proportional to $\cos^2 \theta$, and as R_1 and V_1 are proportional to $\cos \theta$, then each term should likewise be a function of $\cos^2 \theta$. Using this argument and incorporating the trigonometric identity $\cos^2 \theta = \frac{1}{2} \cos(2\theta) + \frac{1}{2}$ demonstrates that V_2 should be of the form

$$V_2 = \hat{V}_2(r_0) e^{i\omega t_0} \cos(2\theta) + \tilde{V}_2(r_0) e^{-i\omega t_0} \cos(2\theta) + \hat{U}_2(r_0) e^{i\omega t_0} + \tilde{U}_2(r_0) e^{-i\omega t_0} \quad (3.38)$$

Substituting this form of the second order velocity into the domain equation, Equation 3.35, and noting that terms that contain $\cos(2\theta)$ are independent of those that do not, give rise to the following equations for $\hat{V}_2$ and $\hat{U}_2$

$$\frac{\partial^2 \hat{V}_2}{\partial r_0^2} + \frac{1}{r_0} \frac{\partial \hat{V}_2}{\partial r_0} - \left(\frac{4}{r_0^2} + \frac{i\omega}{v} \right) \hat{V}_2 = 0 \quad (3.39)$$

$$\frac{\partial^2 \hat{U}_2}{\partial r_0^2} + \frac{1}{r_0} \frac{\partial \hat{U}_2}{\partial r_0} - \frac{i\omega}{v} \hat{U}_2 = 0 \quad (3.40)$$

The solutions to these equations are

$$\hat{V}_2 = k_{21} J_2(i\lambda r_0) + k_{22} Y_2(i\lambda r_0) \quad (3.41)$$

$$\hat{U}_2 = a_{21} J_0(i\lambda r_0) + a_{22} Y_0(i\lambda r_0) \quad (3.42)$$

where the constants are again given in Appendix C. Using the same method, it can be easily shown that the ‘tilde’ equations are

$$\tilde{V}_2 = k_{23} J_2(\lambda r_0) + k_{24} Y_2(\lambda r_0) \quad (3.43)$$

$$\tilde{U}_2 = a_{23} J_0(\lambda r_0) + a_{24} Y_0(\lambda r_0) \quad (3.44)$$

Higher order velocity perturbations can be found, but this study seeks the leading order effects of the eccentricity that only requires velocity and concentration perturbations to the second order.

3.3.2 Perturbed Concentration Equations

As has been done previously, the concentration will be found using the species conservation equations and approximating the concentration with an axial part and a part that depends on the radius, time, and in the eccentric case, θ . As with the velocity, the concentration can be written as a Taylor series similar to Equation 3.20 where C_0 , C_1 , and C_2 would replace V_0 , V_1 , and V_2 respectively. In this section, the species conservation domain equations will be presented for each order of ε along with the appropriate boundary conditions that account for the mapping. The solutions to the zeroth, first, and

second order concentration perturbations will be given with the constants associated with the homogenous solutions reserved for Appendix C.

Order ϵ^0

The species domain equation for the zeroth order perturbation on concentration is independent of the angle, θ , and can be written as

$$\frac{\partial C_0}{\partial t_0} + V_0 \frac{\Delta C}{L} = D \left[\frac{\partial^2 C_0}{\partial r_0^2} + \frac{1}{r_0} \frac{\partial C_0}{\partial r_0} \right] \quad (3.45)$$

where the constant $\frac{\Delta C}{L}$ is $\frac{\partial C_0}{\partial z_0}$ given our definition for the concentration. The boundary conditions are again that the walls of the inner and outer tube are impermeable to each species written as

$$\frac{\partial C_0}{\partial r_0} = 0 \text{ at } r_0 = R_{\text{out}} \text{ and at } r_0 = R_0 \quad (3.46, 3.47)$$

With the concentration written in the form

$$C_0 = \hat{C}_0(r_0)e^{i\omega t_0} + \tilde{C}_0(r_0)e^{-i\omega t_0} \quad (3.48)$$

the solution for the zeroth order concentration is

$$\hat{C}_0 = m_{01}J_0(i\alpha r_0) + m_{02}Y_0(i\alpha r_0) + \frac{\Delta C}{DL} \frac{k_{01}}{\lambda^2 - \alpha^2} J_0(i\lambda r_0) + \frac{\Delta C}{DL} \frac{k_{02}}{\lambda^2 - \alpha^2} Y_0(i\lambda r_0) \quad (3.49)$$

$$\tilde{C}_0 = m_{03}J_0(\alpha r_0) + m_{04}Y_0(\alpha r_0) + \frac{\Delta C}{DL} \frac{k_{03}}{\lambda^2 - \alpha^2} J_0(\lambda r_0) + \frac{\Delta C}{DL} \frac{k_{04}}{\lambda^2 - \alpha^2} Y_0(\lambda r_0) \quad (3.50)$$

where α is the same as previously defined in Equation 1.17. For the next two orders of concentration, only the ‘tilde’ equations will be presented, as they are ultimately the only expressions needed for the mass transfer.

Order ϵ^1

The domain equations for the first order perturbation on the concentration are

$$\frac{\partial C_1}{\partial r_0} + V_1 \frac{\Delta C}{L} = D \left[\frac{\partial^2 C_1}{\partial r_0^2} + \frac{1}{r_0} \frac{\partial C_1}{\partial r_0} + \frac{1}{r_0^2} \frac{\partial^2 C_1}{\partial \theta^2} \right] \quad (3.51)$$

with the boundary equations being

$$\frac{\partial C_1}{\partial r_0} = 0 \text{ at } r_0 = R_{\text{out}} \quad (3.52)$$

and

$$\frac{\partial C_1}{\partial r_0} + R_1 \frac{\partial^2 C_0}{\partial r_0^2} = 0 \text{ at } r_0 = R_0 \quad (3.53)$$

Writing C_1 as

$$C_1 = \hat{C}_1 e^{i\alpha r_0} \cos \theta + \tilde{C}_1 e^{-i\alpha r_0} \cos \theta \quad (3.54)$$

gives the solution

$$\tilde{C}_1 = m_{13} J_1(\alpha r_0) + m_{14} Y_1(\alpha r_0) + \frac{\Delta C}{DL} \frac{k_{13}}{\alpha^2 - \lambda^2} J_1(\lambda r_0) + \frac{\Delta C}{DL} \frac{k_{14}}{\alpha^2 - \lambda^2} Y_1(\lambda r_0) \quad (3.55)$$

Order ϵ^2

Finally, for the second order concentration term, the domain equation is in the same form as the first order concentration equation, or

$$\frac{\partial C_2}{\partial r_0} + V_2 \frac{\Delta C}{L} = D \left[\frac{\partial^2 C_2}{\partial r_0^2} + \frac{1}{r_0} \frac{\partial C_2}{\partial r_0} + \frac{1}{r_0^2} \frac{\partial^2 C_2}{\partial \theta^2} \right] \quad (3.56)$$

The boundary conditions are *

$$\frac{\partial C_2}{\partial r_0} = 0 \text{ at } r_0 = R_{\text{out}} \quad (3.57)$$

and

$$\frac{\partial C_2}{\partial r_0} + \frac{\partial^3 C_0}{\partial r_0^3} R_1^2 + 2 \frac{\partial^2 C_1}{\partial r_0^2} R_1 + \frac{\partial^2 C_0}{\partial r_0^2} R_2 = 0 \text{ at } r_0 = R_0 \quad (3.58)$$

The second order concentration is written in a similar way as the second order velocity found in Equation 3.38

$$C_2 = \hat{C}_2(r_0)e^{i\alpha r_0} \cos(2\theta) + \tilde{C}_2(r_0)e^{-i\alpha r_0} \cos(2\theta) + \hat{S}_2(r_0)e^{i\alpha r_0} + \tilde{S}_2(r_0)e^{-i\alpha r_0} \quad (3.59)$$

This again gives two equations that correspond to terms involving $\cos(2\theta)$ and terms that are independent of θ to give

$$\frac{\partial \tilde{C}_2}{\partial r_0^2} + \frac{1}{r_0} \frac{\partial \tilde{C}_2}{\partial r_0} + \left(\alpha^2 - \frac{4}{r_0^2} \right) \tilde{C}_2 = \frac{\Delta C}{DL} [k_{23} J_2(\lambda r_0) + k_{24} Y_2(\lambda r_0)] \quad (3.60)$$

and

$$\frac{\partial \tilde{S}_2}{\partial r_0^2} + \frac{1}{r_0} \frac{\partial \tilde{S}_2}{\partial r_0} + \alpha^2 \tilde{S}_2 = \frac{\Delta C}{DL} [a_{23} J_0(\lambda r_0) + a_{24} Y_0(\lambda r_0)] \quad (3.61)$$

for the ‘tilde’ terms. The solutions are calculated to be

$$\tilde{C}_2 = m_{23} J_2(\alpha r_0) + m_{24} Y_2(\alpha r_0) + \frac{\Delta C}{DL} \frac{k_{23}}{\alpha^2 - \lambda^2} J_2(\lambda r_0) + \frac{\Delta C}{DL} \frac{k_{24}}{\alpha^2 - \lambda^2} Y_2(\lambda r_0) \quad (3.62)$$

$$\tilde{S}_2 = b_{23} J_0(\alpha r_0) + b_{24} Y_0(\alpha r_0) + \frac{\Delta C}{DL} \frac{a_{23}}{\alpha^2 - \lambda^2} J_0(\lambda r_0) + \frac{\Delta C}{DL} \frac{a_{24}}{\alpha^2 - \lambda^2} Y_0(\lambda r_0) \quad (3.63)$$

As will be evident shortly, only $\tilde{S}_2$ needs to be used in the final calculation for the mass transfer, so only the constants associated with $\tilde{S}_2$ will be given in Appendix C. These solutions represent the perturbations on the velocity and the concentration that will be used to calculate the effect of eccentricity on the mass transfer, and it is the effect of the perturbation on the mass transfer that will be discussed next.

3.3.3 Perturbed Total Mass Transfer

The mass transfer is a scalar quantity defined as the molar flux of each species averaged over the area of interest. For the annular problem, the total mass transfer is

$$Q = \int_{R_{in}}^{R_{out}} \int_0^{2\pi} \left(-D \frac{\partial C}{\partial z} + V_z C \right) r dr d\theta \quad (3.64)$$

which is the sum of the diffusive mass transfer and that due to the convection. If the system is perturbed, like the eccentric system, the mass transfer is also a function of the perturbation in the same way as the velocity and concentration.

$$Q = Q_0 + \varepsilon Q_1 + \frac{1}{2} \varepsilon^2 Q_2 + \dots \quad (3.65)$$

where

$$Q_1 = \left. \frac{dQ}{d\varepsilon} \right|_{\varepsilon=0} = \frac{d}{d\varepsilon} \left[\int_{R_{in}(\varepsilon)}^{R_{out}} \int_0^{2\pi} V_z(\varepsilon, r', \theta') C(\varepsilon, r', \theta') r' dr' d\theta' \right]_{\varepsilon=0} \quad (3.66)$$

and

$$Q_2 = \left. \frac{d^2 Q}{d\varepsilon^2} \right|_{\varepsilon=0} = \frac{d^2}{d\varepsilon^2} \left[\int_{R_{in}(\varepsilon)}^{R_{out}} \int_0^{2\pi} V_z(\varepsilon, r', \theta') C(\varepsilon, r', \theta') r' dr' d\theta' \right]_{\varepsilon=0} \quad (3.67)$$

taking only the *convective* term. A problem arises in taking the total derivative of the integral because one of the limits of integration is a function of ε in $R_{in}(\varepsilon)$. Employing Leibnitz's rule will aid in overcoming this problem so that the mass transfer can be calculated. For simplicity, the argument in the integration will be defined as

$$\psi(\varepsilon, r, \theta) = V_z(\varepsilon, r, \theta) C(\varepsilon, r, \theta) \quad (3.68)$$

ψ also can be perturbed as

$$\psi(\varepsilon, r, \theta) = \psi_0 + \varepsilon \left(\psi_1 + r_1 \frac{\partial \psi_0}{\partial r_0} \right) + \frac{1}{2} \varepsilon^2 \left(\psi_2 + \frac{\partial^2 \psi_0}{\partial r_0^2} r_1^2 + 2 \frac{\partial \psi_1}{\partial r_0} + \frac{\partial \psi_0}{\partial r_0} r_2 \right) + \dots \quad (3.69)$$

where

$$\psi_0 = (V_z C)_0 = V_0 C_0 \quad (3.70)$$

$$\psi_1 = (V_z C)_1 = V_1 C_0 + V_0 C_1 \quad (3.71)$$

and

$$\psi_2 = (V_z C)_2 = V_2 C_0 + V_1 C_1 + V_0 C_2 \quad (3.72)$$

Leibnitz's rule dictates that

$$\frac{dQ}{d\varepsilon} = -\int_0^{2\pi} \frac{dR_m(\varepsilon)}{d\varepsilon} \psi(\varepsilon, R_m(\varepsilon), \theta') R_m(\varepsilon) d\theta' + \int_0^{2\pi} \int_{R_m(\varepsilon)}^{R_{out}} \frac{\partial \psi(\varepsilon, r', \theta')}{\partial \varepsilon} r' dr' d\theta' \quad (3.73)$$

Evaluated at $\varepsilon = 0$, Q_1 becomes

$$Q_1 = \left. \frac{dQ}{d\varepsilon} \right|_{\varepsilon=0} = -\int_0^{2\pi} R_1 \psi_0 R_0 + \int_0^{2\pi} \int_{R_0}^{R_{out}} \psi_1 r' dr' d\theta' \quad (3.74)$$

As can be seen, ψ_1 goes as $\cos \theta$ and R_1 is $\cos \theta$. When this is integrated over θ from 0 to 2π , each term is zero making Q_1 zero. For this problem, the effect of the eccentricity will not be seen to the first order, thus the second order term needs to be found to find the initial effect of the eccentricity.

For the second order perturbation on the mass transfer, Leibnitz's rule is used again to give

$$\begin{aligned} \frac{d^2 Q}{d\varepsilon^2} = & -\int_0^{2\pi} \left\{ \frac{d^2 R_m(\varepsilon)}{d\varepsilon^2} \psi(\varepsilon, R_m(\varepsilon), \theta) R_m(\varepsilon) + \frac{dR_m(\varepsilon)}{d\varepsilon} \left[R_m(\varepsilon) \frac{d\psi(\varepsilon, R_m(\varepsilon), \theta)}{d\varepsilon} \right. \right. \\ & \left. \left. + \psi(\varepsilon, R_m(\varepsilon), \theta) \frac{dR_m(\varepsilon)}{d\varepsilon} \right] \right\} d\theta' \\ & - \int_0^{2\pi} \frac{dR_m(\varepsilon)}{d\varepsilon} \frac{\partial \psi(\varepsilon, R_m(\varepsilon), \theta)}{\partial \varepsilon} R_m(\varepsilon) d\theta' + \int_0^{2\pi} \int_{R_m(\varepsilon)}^{R_{out}} \frac{\partial^2 \psi(\varepsilon, R_m(\varepsilon), \theta)}{\partial \varepsilon^2} r' dr' d\theta' \end{aligned} \quad (3.75)$$

Evaluating this at $\varepsilon = 0$ and recalling that

$$\frac{d\psi}{d\varepsilon} = \psi_1 + r_1 \frac{d\psi_0}{dr_0} \quad (3.76)$$

gives for the second order mass transfer perturbation

$$\begin{aligned} Q_2 = & -\int_0^{2\pi} R_2 \psi_0 R_0 d\theta' - \int_0^{2\pi} R_1 R_0 \left(\psi_1 + R_1 \frac{d\psi_0}{dr_0} \right) d\theta' - \int_0^{2\pi} \psi_0 R_1^2 d\theta' \\ & - \int_0^{2\pi} R_1 \psi_1 R_0 d\theta' + \int_0^{2\pi} \int_{R_0}^{R_{out}} \psi_2 r' dr' d\theta' \end{aligned} \quad (3.77)$$

Next, the equations for the velocity and concentration given earlier will be used for ψ_0 , ψ_1 , and ψ_2 . As has been done previously, the mass transfer will be time averaged over one period ($2\pi/\omega$). Finally the expressions for R_1 , and R_2 will be substituted into the expression, and the ' θ ' integrals are calculated to give for the perturbed mass transfer

$$\begin{aligned} \overline{Q}_2 = & -2\pi R_0 \left[2\Re(\hat{V}_0 \tilde{C}_1) + 2\Re(\hat{V}_1 \tilde{C}_0) + \Re \left\{ \frac{d(\hat{V}_0 \tilde{C}_0)}{dr_0} \right\} \right] \\ & + 2\pi \int_{R_0}^{R_{out}} \left[2\Re(\hat{V}_0 \tilde{S}_2) + \Re(\hat{V}_1 \tilde{C}_1) + 2\Re(\hat{U}_2 \tilde{C}_0) r' dr' \right] \end{aligned} \quad (3.78)$$

This is the final expression used for the perturbed mass transfer that will be used in the subsequent analysis. Keep in mind that $\overline{Q}_0$ involves the diffusive and convective mass transfer, but because the diffusive term is unaffected by the perturbation, $\overline{Q}_0 + \frac{1}{2}\epsilon^2 \overline{Q}_2$ is the initial effect of the perturbation on the total mass transfer.

3.3.4 Results and Analysis

Maple® is the software package used to calculate Equation 3.78 and produce graphs of the mass transfer versus the frequency of oscillation. The second order perturbation on the mass transfer involves numerous forms of the Bessel function in a complicated way. Therefore, Maple® is unable to calculate $\overline{Q}_2$ for large values of the argument in the Bessel functions. The parameters given in Table 3.1 are chosen so that this limitation of the software would be delayed until larger values of ω and a reasonable

analysis of the results could be made before the problems began. With this brief preface, the results will now be given.

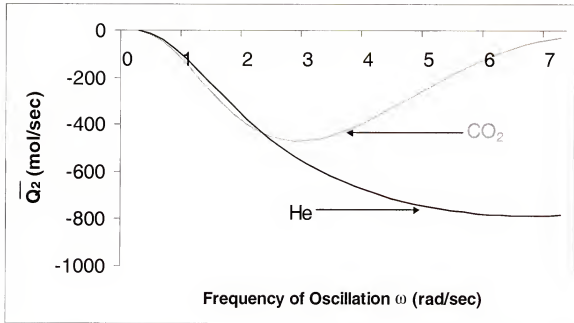


Figure 3.10 Curves for the second order perturbation of the mass transfer versus frequency for carbon dioxide and helium.

Fig. 3.10 is a plot of the second order perturbation on the mass transfer versus the frequency of oscillation for two species. As can be seen, the second order perturbation is initially negative and decreasing and then begins to increase towards zero. This demonstrates that the eccentricity initially decreases the total mass transfer. If the inner tube is slightly off center, this modifies the flow in the annular region and causes the total mass transfer to become less. This can be explained physically.

One can see that when the inner cylinder is shifted, there is portion of the annulus that has become “thicker” and a portion that has become “thinner”. Appendix D shows that there is more of the portion that is now “thicker” and less that is “thinner”. In thinking back to oscillating flat plates, there is a slow flow region that exists in the core

of the fluid away from the fast moving boundary. If a species diffuses into the core of slow flow, it will hinder its mass transfer more than if remained near the boundary. If the distance between the flat plates is increased slightly and all other parameters remain unchanged, the region of slow flow has also increased. This will hinder the mass transfer of a species more because there is a larger slow flow region. The same is true in an annulus. The “thicker” portion of the annulus has a larger region of slow flow in comparison to the centered inner cylinder. This will cause the mass transfer to decrease as this “thicker” portion dominates because there is more of it.

At large frequencies, most of the radial movement of species will be confined to a small region close to the moving boundary as each new cycle begins very rapidly. As the movement of the diffusing species remains very near the boundary, the influence of one moving boundary on the other is very small. For an annulus at high frequencies, it therefore should not matter whether the inner tube is centered or slightly. Although not shown in Fig. 3.10 due to the limitation of Maple® already discussed, the second order perturbation should tend towards zero for high frequencies. However, there is a range of frequencies between the two extremes discussed above where the frequency is large enough to limit the effect in the “thicker” portion of the annulus but still retain the effect of the “thinner” portion. If the walls of the annulus are brought close together, the region of slow flow shrinks resulting in an increase in the mass transfer. This causes $\overline{Q}_2$ to become less negative as shown in Fig. 3.10 for moderate frequencies. The effect of eccentricity on the mass transfer for species with differing diffusion coefficients requires a look into the size of the viscous and diffusive boundary layers of the system.

As the frequency increases, the viscous boundary layer, or Stokes layer, becomes thinner for increasing frequency according to the relation

$$\text{Stokes Layer} = \sqrt{\frac{\nu}{\omega}} \quad (3.79)$$

The size of the Stokes layer is the same for He and CO₂ since it is a measure of the viscous boundary layer of the oscillating medium, nitrogen. The diffusive boundary layer is then the square root of the ratio of the diffusion coefficient for each species to the frequency, or

$$\text{Diffusive Boundary Layer} = \sqrt{\frac{D_i}{\omega}} \quad (3.80)$$

where i is either He or CO₂. It can be readily seen that the diffusive boundary layer for He is larger than that for CO₂ because the diffusion coefficient for He is about 4.3 times that of carbon dioxide. For CO₂ the viscous and diffusive boundary layer are approximately the same size, but the diffusive boundary layer for He is about 2.1 times the size of the stokes layer of the fluid at all frequencies. This explains why the eccentricity negatively affects the mass transfer for a faster diffuser more than a slower diffuser. As the region of slow flow increases in the thicker portion of the eccentric annulus, the faster diffuser moves more into this region than the slower diffuser. As stated earlier, as the frequency increases, most of the mass transfer takes place near the boundary of the tubes as the viscous and diffusive boundary layer both decrease. However, this affects the faster diffuser more slowly than the slow diffuser. That is why the minimum in the perturbed mass transfer for CO₂ occurs before the minimum for He. In fact, the minimum for He occurs at a frequency 2.1 times higher than for CO₂ since this is the numerical ratio of the diffusive boundary layer for each species. The relative

locations for the minimum in the perturbed mass transfer of both species are verified in Fig. 3.10. Because the eccentricity affects each species differently, this will have an effect of the separation that can be achieved in an eccentric geometry.

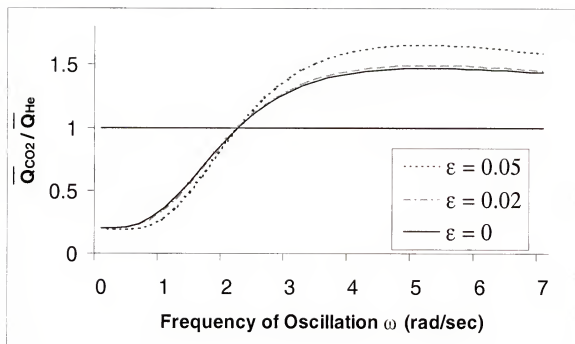


Figure 3.11 Separation ratio for a centered annulus ($\epsilon = 0$) and configurations where the inner cylinder has been displaced by two different amounts.

As has been done before, the ratio of the mass transfer for two different species will show the separation that can be achieved in periodic annular flow. Fig. 3.11 demonstrates the separation that can be achieved for a CO_2 -He system in a nitrogen carrier for a centered annulus and an eccentric annulus displaced by separate amounts. The value of ϵ is chosen so that it is large enough to give a noticeable difference between the perturbed and unperturbed case yet not too large to discount the Taylor series expansion about small ϵ that is the basis for the perturbation analysis. As can be seen in the ratio of the slow diffusing CO_2 to the faster diffusing He, the separation of species

increases for an eccentric geometry and continues to increase for greater displacements. This is true if one was to separate out He first as the ratio of mass transfers is more of a fraction for the eccentric annulus, and it is also true for the separation of more CO₂ as the ratio is higher for the new geometry.

Although an eccentric annulus in periodic flow will decrease the mass transfer for each species in comparison to the centered inner cylinder, it does enhance the separation achieved in a binary system. This information will prove useful for those that use this method in separation processes and in the analysis of the experiments performed on annular flow that will be presented later in this thesis.

CHAPTER 4 MORE ON PERIODIC ANNULAR FLOW

This chapter will continue the analysis of periodic annular flow. This chapter will consider oscillating out of phase and tubes oscillating in phase at different amplitudes in boundary driven flow. Instead of using the circular geometry, flow in a channel will again be utilized as a simplified model to observe qualitative results and the physics associated with these new changes. The previous chapters have shown that the trends observed for the two-dimensional case continue to be true for the circular geometries. Therefore, by having plates oscillate out of phase and having one plate oscillate at a different amplitude, these configurations will be analyzed to demonstrate qualitatively what may happen to the mass transfer and separation during an experiment in periodic annular flow.

4.1 Recalculation of the Velocity, Concentration, and Mass Transfer Profiles

4.1.1 Plates Oscillating with Phase Angle ϕ

The equation of motion for boundary driven flow in a two-dimensional channel is again

$$\frac{\partial V_x}{\partial t} = \nu \frac{\partial^2 V_x}{\partial y^2} \quad (4.1)$$

with boundary conditions

$$V_x = \frac{1}{2} A \omega \cos(\omega t) \text{ at } y = h \quad (4.2)$$

and

$$V_x = \frac{1}{2} A \omega \cos(\omega t + \phi) \text{ at } y = 0 \quad (4.3)$$

indicating that the bottom plate oscillates at a phase angle, ϕ , different than the upper plate. The phase angle can range from $0 \leq \phi \leq \pi$ (0 to 180°) because over a cycle it will not matter if the bottom plate lags behind the upper plate or oscillates ahead of the upper plate. Letting

$$V_x = \hat{V}_x e^{i\omega t} + \tilde{V}_x e^{-i\omega t} \quad (4.4)$$

the unscaled solution for the velocity is

$$\hat{V}_x = \frac{1}{4} A \omega \frac{e^{i\phi - \lambda h} - 1}{e^{-\lambda h} - e^{\lambda h}} e^{\lambda y} + \frac{1}{4} A \omega \frac{1 - e^{i\phi + \lambda h}}{e^{-\lambda h} - e^{\lambda h}} e^{-\lambda y} \quad (4.5)$$

and

$$\tilde{V}_x = \frac{1}{4} A \omega \frac{e^{-i(\phi + \lambda h)} - 1}{e^{-i\lambda h} - e^{i\lambda h}} e^{i\lambda y} + \frac{1}{4} A \omega \frac{1 - e^{i(\lambda h - \phi)}}{e^{-i\lambda h} - e^{i\lambda h}} e^{-i\lambda y} \quad (4.6)$$

for tubes oscillating out of phase. The expression for λ is the same as in Equation 1.7.

The species conservation equation will be the same as Equation 2.6, and the solution for $\hat{c}$ with the new velocity profile given above is now

$$\begin{aligned} \hat{c} = & \frac{(\hat{k}_2 - \hat{k}_1) e^{-\alpha h} + \hat{k}_1 e^{\lambda h} - \hat{k}_2 e^{-\lambda h}}{(\lambda^2 - \alpha^2)(e^{-\alpha h} - e^{\alpha h})} \frac{\lambda}{\alpha} e^{\alpha y} + \frac{(\hat{k}_2 - \hat{k}_1) e^{\alpha h} + \hat{k}_1 e^{\lambda h} - \hat{k}_2 e^{-\lambda h}}{(\lambda^2 - \alpha^2)(e^{-\alpha h} - e^{\alpha h})} \frac{\lambda}{\alpha} e^{-\alpha y} \\ & + \frac{\hat{k}_1 e^{\lambda y} + \hat{k}_2 e^{-\lambda y}}{\lambda^2 - \alpha^2} \end{aligned} \quad (4.7)$$

where

$$\hat{k}_1 = \frac{\frac{1}{4} A \omega (c_2 - c_1)}{DL} \frac{e^{i\phi - \lambda h} - 1}{e^{-\lambda h} - e^{\lambda h}} \quad (4.8)$$

and

$$\hat{k}_2 = \frac{\frac{1}{4} A \omega (c_2 - c_1)}{DL} \frac{1 - e^{i\phi + \lambda h}}{e^{-\lambda h} - e^{\lambda h}} \quad (4.9)$$

The total time- and space-averaged mass transfer is again calculated by using

Equation 2.11. Letting

$$\tilde{V}_x = \frac{1}{4} A \omega (\psi_1 e^{\beta \lambda y} + \psi_2 e^{-\beta \lambda y}) \quad (4.10)$$

and

$$\hat{c} = \frac{1}{4} A \frac{(c_2 - c_1)}{L} (\phi_1 e^{\alpha y} + \phi_2 e^{-\alpha y} + \phi_3 e^{\lambda y} + \phi_4 e^{-\lambda y}) \quad (4.11)$$

where the ψ 's and ϕ 's can be determined by inspection of the above equations. The final expression for the mass transfer is the unscaled form of Equation 2.37 except now the values for the ψ 's and ϕ 's replace the β 's and η 's respectively. In unscaled form,

Equation 2.37 with the accounted phase shift is now

$$\bar{Q} = -\frac{D(c_2 - c_1)}{L} + \frac{A^2 \omega (c_2 - c_1)}{8Lh} \Re \left[\begin{aligned} & \frac{\psi_1 \phi_1 (e^{(i\lambda - \alpha)h} - 1) - \psi_2 \phi_2 (e^{(\alpha - i\lambda)h} - 1)}{(i\lambda - \alpha)} \\ & + \frac{\psi_1 \phi_2 (e^{(i\lambda + \alpha)h} - 1) - \psi_2 \phi_1 (e^{-(i\lambda + \alpha)h} - 1)}{(i\lambda + \alpha)} \\ & + \frac{\psi_1 \phi_3 (e^{(i+1)\lambda h} - 1) - \psi_2 \phi_4 (e^{-(i+1)\lambda h} - 1)}{(i+1)\lambda} \\ & + \frac{\psi_1 \phi_4 (e^{(i-1)\lambda h} - 1) - \psi_2 \phi_3 (e^{-(i-1)\lambda h} - 1)}{(i-1)\lambda} \end{aligned} \right] \quad (4.12)$$

4.1.2 Plates Oscillating in Phase but at Different Amplitudes

Moving on to the second case, the upper and lower plate are assumed to now oscillate in phase but at different amplitudes. The equation of motion is the same, but the boundary conditions now become

$$V_x = \frac{1}{2} A_1 \omega \cos(\omega t) \text{ at } y = h \quad (4.13)$$

and

$$V_x = \frac{1}{2} A_2 \omega \cos(\omega t) \text{ at } y = 0 \quad (4.14)$$

For this configuration, the velocity is given via

$$\hat{V}_x = \frac{1}{4}\omega \frac{A_2 e^{\lambda h} - A_1}{e^{-\lambda h} - e^{\lambda h}} e^{\lambda y} + \frac{1}{4}\omega \frac{A_1 - A_2 e^{\lambda h}}{e^{-\lambda h} - e^{\lambda h}} e^{-\lambda y} \quad (4.15)$$

and

$$\tilde{V}_x = \frac{1}{4}\omega \frac{A_2 e^{-i\lambda h} - A_1}{e^{-i\lambda h} - e^{i\lambda h}} e^{i\lambda y} + \frac{1}{4}\omega \frac{A_1 - A_2 e^{i\lambda h}}{e^{-i\lambda h} - e^{i\lambda h}} e^{-i\lambda y} \quad (4.16)$$

The concentration is of the same form as Equation 4.7, but the different velocity profile makes

$$\hat{k}_1 = \frac{\frac{1}{4}\omega(c_2 - c_1)}{DL} \frac{A_2 e^{\lambda h} - A_1}{e^{-\lambda h} - e^{\lambda h}} \quad (4.17)$$

and

$$\hat{k}_2 = \frac{\frac{1}{4}A\omega(c_2 - c_1)}{DL} \frac{A_1 - A_2 e^{\lambda h}}{e^{-\lambda h} - e^{\lambda h}} \quad (4.18)$$

for the present configuration. The velocity and concentration can again be written in the same form as Equation 4.10 and 4.11 respectively resulting in the total time- and space-averaged mass transfer given in Equation 4.12. The only difference in Equation 4.12 for the plates oscillating out of phase and for this configuration is again in the ψ 's and φ 's that can be determined by inspection of the velocity and concentration profiles for this case.

4.2 Results and Discussion

4.2.1 Plates Oscillating With Phase Angle ϕ

Fig. 4.1 shows the total mass transfer of Helium in a Nitrogen carrier for increasing frequency at two phase angles. The reference curve in Fig.4.1 is a

configuration where both plates are oscillating in phase ($\phi = 0$) and the second curve represents both plates are 180° out phase ($\phi = \pi$). For low frequencies, the configuration

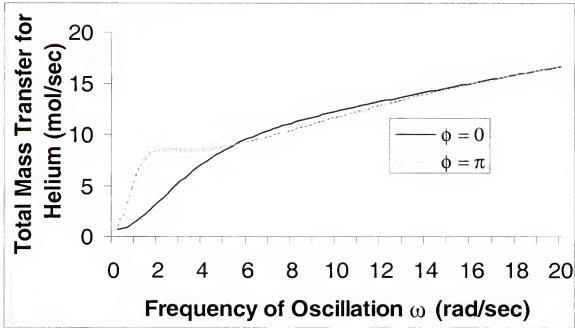


Figure 4.1 Total mass transfer of helium for plates oscillating in phase ($\phi = 0$) and 180° out of phase ($\phi = \pi$).

with the plates out of phase has a higher mass transfer than the reference configuration.

Although this plot is specifically for a phase angle of 180° , it can be shown that any phase angle greater than zero will give a higher mass transfer for small frequencies. For high frequencies the mass transfer favors plates oscillating in phase as seen in Fig. 4.1.

Finally, for even higher frequencies, there is little difference between the two configurations as the mass transfer is essentially the same. A physical explanation will be offered to account for the phenomena observed.

The explanation continues to focus on a species' ability to remain in the faster moving regions of the flow field in the direction of the concentration gradient. For plates

oscillating *in phase*, the fast moving region is near the boundary of the two plates with the slow moving region being in the middle. For plates oscillating *out of phase*, imagine that at some time during the oscillations, the lower plate has moved as far as it can in the

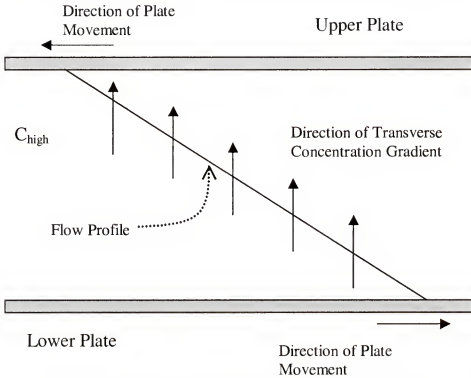


Figure 4.2 Depiction of flow profile and concentration gradient for plates oscillating out of phase.

direction of the axial concentration gradient and the upper plate is lagging 180° behind it as seen in Fig. 4.2. Because of the flow profile, the transverse concentration gradients will be unidirectional from the lower plate to the upper plate. At that instant, the upper plate is currently moving in the opposite direction of the desired mass transfer. However, as the species begins to diffuse toward the upper plate, the upper plate begins to move in the direction of the desired mass transfer and begins to convect the species down the channel. It appears that the species is consistently taking advantage of the fluid movement in the desired direction of mass transfer and not becoming stagnant in the

middle of the channel for the case of plates oscillating in phase. In fact, a type of resonance seems to occur where the transverse diffusion of the dilute species and the forward motion of the fluid are occurring at an optimum rate. This is seen in Fig. 4.1 by the “hump” in the curve occurring at a frequency of approximately 2 rad/sec.

As the frequency increases, the dilute species is unable to diffuse as far in the transverse direction in the time it takes to complete a cycle. At higher frequencies, the slow moving region in between the plates becomes even *more* stagnant for plates oscillating out of phase than for the configuration of plates oscillating in phase. This will then hinder the mass transfer more for the out of phase case as seen in Fig.4.1 for higher frequencies.

Finally, for very high frequencies, the Stokes layer where most of the mass transfer occurs becomes very small for both configurations. Similar to the argument for an eccentric annulus, at very high frequencies, the lower plate is virtually unaware of the existence of the upper plate and vice versa. Therefore, it will not make a difference whether one plate is oscillating in phase or out of phase with the other. This accounts for the mass transfer being essentially the same for both configurations at very high frequencies in Fig. 4.1.

The separation ratio for a CO₂-He binary system for two phase angles is given in Fig. 4.3. As can be seen, the curves change dramatically for different phase angles. For example, a large separation can be achieved around 2 rad/sec for both configurations. For plates oscillating in phase, CO₂ will separate out more at this frequency; however, for plates oscillating 180° out of phase, exactly the opposite occurs as more helium will separate out at 2 rad/sec. It can be seen that for a high throughput, plates oscillating *in*

phase will give a greater separation for CO₂ and plates oscillating 180° *out of phase* will give a better separation of the faster diffusing helium.

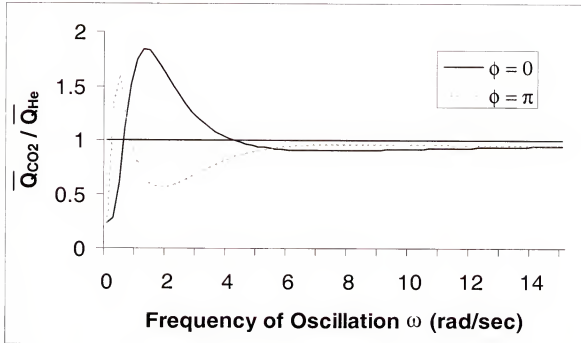


Figure 4.3 Separation ratio for a carbon dioxide and helium system for plates oscillating in phase and out of phase.

4.2.2 Plates Oscillating in Phase but at Different Amplitudes

The explanation of the physics for plates oscillating at different amplitudes is straightforward given the explanation of the physics behind periodic flow in Chapter 2. For example, imagine two plates oscillating at a peak-to-peak amplitude of 10 cm. If the amplitude of one of the plates is increased to 20 cm, the overall velocity available for transport has also increased. The opposite would be true if the amplitude of one of the plates decreases to 5 cm. One would expect that the total mass transfer would increase for an increase in one of the amplitudes and decrease if the amplitude is made smaller.

This is seen in Fig. 4.4 showing the total mass transfer of Helium for three configurations where the amplitudes are the same, one is larger, and one is smaller.

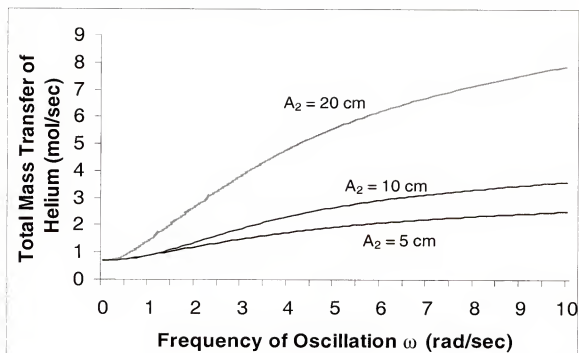


Figure 4.4 Total mass transfer of helium versus frequency for three different values for the amplitude of one of the plates.

As it is the convection of the carrier fluid that allows a slow diffusing species to have a higher transport than a fast diffusing species, the separation would be greater for a configuration with a higher overall velocity. The disadvantage a slower diffusing species initially has because of its lower diffusion coefficient is remedied because of its ability to remain closer to the faster moving boundary than the faster diffusing species at low frequencies. If one of the boundaries is moving faster because of a larger oscillation amplitude, this simply adds to the advantage the slower diffusing has over a faster diffusing species. The reverse is again true if one of the plates operates at a smaller

amplitude. Fig. 4.5 shows the effect of the same three configurations analyzed previously on the separation for a CO_2 -He binary system.

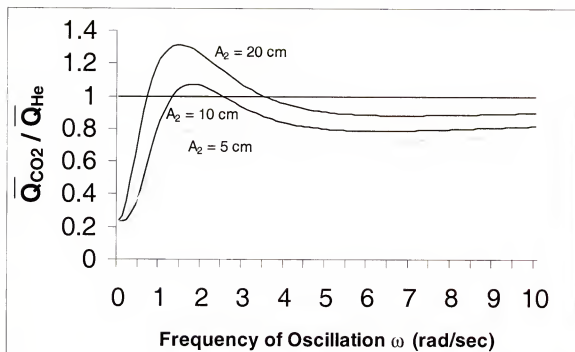


Figure 4.5 Separation ratio for a helium and carbon dioxide system for three different amplitudes for plates oscillating at different amplitudes.

The results of these simple models show that operating plates out of phase or at different amplitudes can affect the mass transfer and separation of dilute species. It is also shown that these changes can result in an advantage in terms of overall transport and in the separation of certain species. Flow through an annular region may give slightly different results because the flow profile is not symmetric as it is in flow in a channel. There would be a difference in the amount by which the mass transfer is affected if the outer tube is oscillating out of phase or at a different amplitude or if the inner tube has the imperfection. However, the trends observed ought to hold true for periodic annular flow. In fact, as the gap width between the two cylinders becomes small compared to the radius

of the inner tube, the results of channel flow and annular flow ought to closely resemble on another. Once again, these findings can be used to help optimize a system to give a reasonable separation at a high throughput.

CHAPTER 5 EXPERIMENTS

Experiments were conducted to help verify the phenomena observed by theory in the previous chapters. The geometries examined experimentally were the oscillating open tube and periodic flow in an annular region. Poplasky has given a detailed description of the apparatus in the experimental section of his thesis [1]. This chapter will give an overview of the apparatus and experimental procedures used to produce the results in this chapter, but if more detail on the apparatus is needed, the reader is referred to the thesis of Poplasky. This chapter will then proceed to give the experimental results for an open tube and an annular geometry. Discussions will then follow on these results comparing them to the model predictions.

5.1 Experimental Apparatus

An overall view of the experimental apparatus is given in Fig.5.1. Periodic flow in this set up was produced using a boundary driven configuration. Motors turn a flywheel at a given angular speed that is then converted into linear motion by oscillating the tubes that connect the reservoirs. If annular periodic flow is to be studied, inner and outer tubes are oscillated by separate motors according to a specified speed and/or a specified phase angle difference between the motion of each tube. If an open tube configuration is desired, the inner tube can be removed giving rise to only an outer tube configuration to produce periodic flow. The amplitude of oscillation can be changed by

altering the position of the "Oscillatory Plate" shown in Fig. 5.1 for each tube along the radius of the flywheel. In order to determine the mass transfer for an experimental run at

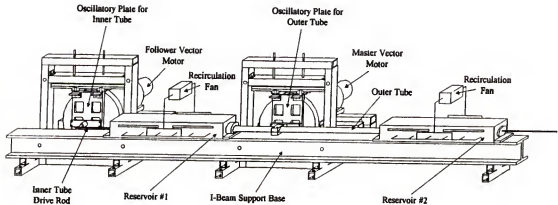


Figure 5.1 Overall view of experimental apparatus.

a specified configuration, the concentration of the species in each reservoir was determined. As both the carrier and dilute species flowed in and out of each reservoir, samples of the concentration for each inlet and outlet stream were then analyzed.

5.1.1 Motion Control

Baldor® Vector Drive Motors were used to drive the oscillatory motion of the system. A computer controlled the angular speed of the motors. The deviations from the desired angular speed that occurred from time to time during an experiment were always within 2% of the set speed. For periodic flow in an annular tube, each tube could act independently by providing each motor with a separate speed, or the motion of one motor could be set to follow the motion of the other. This was important in running experiments for tubes oscillating at the same frequency but with a phase angle ϕ between

them. Having one motor follow the other could be accomplished, as the position of each motor could be determined at anytime during its rotation. An encoder for each motor defined 1024 counts per rotation of each motor. If the Follower motor maintained its relative position to that of Master motor, the starting phase difference between the motors would be kept constant throughout the entire experiment. A Baldor® SmartMove Motion Controller ensured that the Follower motor maintained its position with the Master. A computer code written in the industrial computer language MINT™ supplied the needed information and kept the motors at the desired configuration until changed by the user. An analysis on the encoder counts of each motor was performed to make sure the Follower motor was moving according to the position of the Master. Although the position of the Follower deviated slightly from the Master, the deviation remained more or less the same and did not become progressively worse throughout the experiments. These modifications allowed for complete control of the motors according to their speed and relative position.

5.1.2 Gas Delivery and Control

Because the concentration of each species needed to remain fixed in order to achieve steady state in the total mass transfer and to ensure that no net flow occurred from one reservoir to the other, a highly accurate gas delivery and control method was used to limit the fluctuations in the inlet concentrations and in the overall flow rates into and out of each cylinder. The gas was initially supplied by cylinders containing each desired species in high purity. For a binary system present in a carrier, both dilute species and the nitrogen carrier flowed from the cylinders to a Matheson® Rotameter used to control the relative amounts of each species supplied to Reservoir 1. The mixture

was sent to a static mixer that provided excellent mixing for the gas flow rates used for the experiments. The gases were then fed to the MKS® Mass Flow Controllers (MFC's). The mass flows were converted to readings in SCCM (volumetric flow) by the MFC's. They served as highly accurate flow controllers for both the inlets and the outlets to each reservoir as each MFC was rated to be within 8 SCCM of the desired flow rate. Since experiments were performed with inlet and outlet flow rates of 900 SCCM, the fluctuation accounted for only 0.89% of the desired value. The gases were then fed to Reservoir 1, exited the reservoir to another MFC, and then vented to the atmosphere. For Reservoir 2, pure nitrogen was fed from the cylinder, to the rotameter, straight to its mass flow controller, and then to the reservoir. The gases then exited Reservoir 2 to another MFC and also vented to the atmosphere. Flow information could be monitored by the MKS® 4 Channel Readout and controlled and further monitored by a computer that read and provided information to the 4 Channel Readout through an MKS® RS 232 Interface. This configuration provided reliable control of the overall flow rates into and out of each reservoir and the relative concentration of the species involved in each experiment.

5.1.3 Concentration Measurement

Samples of the gas mixture were taken at the inlet and outlet of Reservoir 1 and the outlet of Reservoir 2 simultaneously using pressure locked gas syringes. Each sample was then analyzed using a Hewlett Packard® 5790 Gas Chromatograph (GC). The GC is a thermal conductivity detector that measures any species having a thermal conductivity different than the carrier gas, usually a Helium or Argon carrier. The GC provided measurements up to a thousandth of a mass percent, and was accurate to within 2-3 % of gas standards and reproducible within 1-2 % using careful sampling and injection

technique. The GC was calibrated periodically, usually before a day's experiments, using standardized gases containing all the species used in the experiments. A Hewlett Packard® 3393A Computing Integrator then recorded the measurements of the GC. The concentration measurement technique employed during these experiments provided highly accurate measurements of the gas mixture composition.

5.1.4 Gas Recirculation

In order to assume the concentration at the outlets of the reservoir to be the same as the concentration throughout the entire reservoir requires each reservoir to be well mixed. Looking again at the experimental setup in Fig. 5.1, note that recirculation fans were used on each reservoir. Both fans blow air through the reservoir in a direction transverse to the tube motion; therefore, this should not appreciably affect the flow profiles in the tube. An experiment was performed to verify the continuous stirred tank model assumed for our experiments. The details of this experiment follow.

In order to initially start the recirculation measurement experiment, the opening in the tube in Reservoir 1 was blocked so that the reservoir was isolated from the rest of the assembly. With the gases stabilized, without any tube motion, and with only the fans turned on, CO₂ was then introduced into the reservoir. After some time, the flow of CO₂ was stopped and measurements began. If the reservoir behaved like a continuous stirred tank, the concentration at the outlet was expected to fall exponentially with time. In fact, after 5 times the residence time of the tank, the concentration of CO₂ in the tank ought to have been 0.7 % of the initial concentration. For a 7.3 L tank with a flow of 900 SCCM, the residence time of the tank is 8.1 min. After 40 min, or five times the residence time, the concentration of CO₂ in the tank was 2.3 % of the initial concentration. This closely

approximates the result one would expect from a continuous stirred tank model and validates the well-mixed approximation.

5.2 Experimental Method

After a few initial experimental runs consisting of observations of the response of the apparatus to various input parameters, a standardized experimental method was devised to maximize the accuracy of each experiment and the reproducibility from run to run. The following is an example of a typical experimental run. It is meant to show the potential for error in each run and where improvements can be made for further experiments in such a study.

To begin the experiment, the nitrogen carrier gas was first introduced into each reservoir at 900 SCCM. This was allowed to equilibrate in each reservoir until the outputs of each reservoir matched the inputs within the error of the MFC's. If the flow rate out of the reservoir was less than it should be, this was an indication of a leak in the assembly. Using an industrial soap solution, leaks were detected and then corrected to maintain a proper mass balance of flow into and out of each reservoir.

After the flows had been equilibrated, the dilute species used for the experiments were then introduced and the recirculation fans turned on. Initially, as seen by the rotameter, the species fluctuated slightly. After some time, the fluctuations died down and a constant concentration is maintained. Before the motors were started, the species were allowed time to reach a steady concentration. Once this was achieved, the motors were started at a desired frequency and, if two tubes were present, a phase angle was set. The phase angle is not a directly inputted parameter in the computer program. The initial starting position of each motor was done visually by moving each flywheel to a desired

starting position. This can be done fairly accurately, but some human error is certainly involved. Once the relative starting positions were set, the phase angle remained the same throughout the experiment.

Approximately 30 minutes was allowed before concentration samples were taken. Samples were taken at the same time from the reservoir in order to provide a “snapshot” of the concentrations at a given moment. An analysis was then done on the samples using the gas chromatograph. Particular attention was made to the concentration measurement in the sample from the outlet of Reservoir 2, as it is this measurement that determines the mass transfer. Once the concentration in this outlet was within five hundredths of a mass percent for three consecutive “snapshots” of concentration taken, the run was considered to have achieved equilibrium, and another run at another frequency was started. Runs at three to five different frequencies were often completed in a day of experiments.

5.3 Sensitivity Analysis

In order to properly compare theoretical calculations with experimental results, the parameters used in the calculations from the experimental setup are subject to error. Error can arise from the accuracy of the instruments used during the experiments, from the precision in the manufacturing of the apparatus, from improper assumptions regarding the experiment, or from human error due to faulty techniques to name a few examples. The following sensitivity analysis is a result of error that can be quantified in order to analyze its effect on the calculation and results. Much of the error on the machining precision is drawn from Poplasky’s [1] thesis, as it is he who assembled the experimental apparatus.

Starting with the motors, it was observed that the motors were within 1 RPM of the set value. This could be as much as 10 % of the desired value at low frequencies (~ 0.2 Hz) or as low as 0.8 % at higher frequencies (~ 2 Hz). However, since the time required for a steady state value to be achieved in the concentration results in the completion of many cycles, the error in the motor frequency should essentially average to the desired value. Further, since the theoretical calculations are time averaged over one cycle, this should also help to minimize the effect of the deviations from the set point. Therefore, the error resulting from the accuracy of the motor control is deemed insignificant as far as the final result is concerned.

For the concentration of the samples analyzed in the gas chromatograph, the chromatograph measured standardized samples to within 2 % of the given value. The accuracy of analysis on the standardized gas from the manufacturer, Scotty®, was rated to be within 5 % of the given composition, thus the gas chromatograph is within the error of the standard sample. Further, steady state was assumed once the outlet concentration to Reservoir 2 was within 5 thousandths of each other over three consecutive samples. Given the relative magnitude of the outlet concentrations (~ 0.1 - 0.2 mass percent) and the fact that these consecutive samples were then averaged, the error in the differences between samples to the outlet concentration to Reservoir 2 was about 2 %. Using the same rationale, the outlet concentration from Reservoir 1 varied within approximately 2 %. Therefore, the total error in concentration measurement was about 4 % for both reservoir outlets.

The sensitivities of the values used for the diffusion coefficients and kinematic viscosity are less straightforward as they involve a number of factors. The diffusion

coefficients for helium, carbon dioxide, and methane in a nitrogen carrier were found in *Tables on the Thermophysical Properties of Liquids and Gases* [24] which gave empirical values for each at different temperatures. It was assumed that the temperature for each experiment was approximately 25° C. According to thermistors that measured the temperature inside each reservoir, the actual temperature fluctuated between 23 and 26° C. According to the *Tables*, the value of the diffusion coefficient varies roughly as $T^{1.7}$. A 2° C fluctuation in the temperature would give a deviation in the diffusion coefficient of approximately 1 % of the given value. The value of diffusion coefficients is also inversely proportional to the pressure [25]. During an experiment, the reservoirs were approximately 2.5 PSI above atmospheric pressure (14.7 PSI). This would make the diffusion coefficient approximately 85 % of the reported value for 1 atmosphere. It is assumed that the daily change in atmospheric pressure is not enough to appreciably affect the value of the diffusion coefficients. Furthermore, the pressure in each tank could fluctuate when the tubes are set in motion, but it is assumed that this fluctuation will have little effect in modifying the value of the diffusion coefficient. The corrected values for the diffusion coefficient used in the model are given in Table 5.1.

Table 5.1 Diffusion coefficients of species in experimental runs

Species	Experimental Value in N ₂ at 298 K and 1 Atm	Actual Value Corrected For Pressure
Carbon Dioxide	0.165 cm ² /sec	0.141 cm ² /sec
Methane	0.215 cm ² /sec	0.184 cm ² /sec
Helium	0.710 cm ² /sec	0.607 cm ² /sec

The kinematic viscosity was determined using the experimental data available [26] for the dynamic viscosity (μ) and density (ρ) of N_2 at 1 atm and 298 K. The dynamic viscosity is a weak function of temperature and pressure; therefore, its value will not change much with varying conditions. The density, however, is directly proportional to the pressure and indirectly proportional to the temperature from the ideal gas relation

$$\rho = \frac{P}{RT} \quad (5.1)$$

The experimental value for the density was adjusted as experiments were again run at 2.5 PSI above atmospheric pressure. The value for the kinematic viscosity of nitrogen is given in Table 5.2. Because the temperature varies about 2 °C and the pressure is accurate to within 0.2 PSI, the error in the kinematic viscosity is +/- 0.7 %.

Table 5.2 Thermophysical parameters of the carrier gas

Parameter	Experimental Value at 1 atm and 298 K	Actual Value Corrected For Pressure
Density (ρ)	0.00112 g/cm ³	0.00131 g/cm ³
Viscosity (μ)	0.000178 g/cm sec	0.000178 g/cm sec
Kinematic Viscosity (ν)	0.159 cm ² /sec	0.136 cm ² /sec

The error in each of the above parameters, along with the error in mechanical precision, will collectively cause an error in the theoretical prediction for the total mass transfer. The amount it alters the mass transfer also depends on the frequency of oscillation. The following table summarizes the error in each of the variables used in the theoretical model and its effect on the calculation of the *effective diffusion coefficient* for an oscillation frequency of 1 Hz (2π rad/sec). Although most of the analysis to this point has been focused on the total mass transfer, the effective diffusion coefficient was used

for the experiments because it normalized the mass transfer with the concentration difference. As it was difficult to maintain the same inlet concentration from day to day and even throughout the day for one experiment, the effective diffusion coefficient helped to standardize the results. The physics observed for the effective diffusion coefficient was the same as the total mass transfer with its definition being

$$D_{eff} = \frac{\bar{Q}_{total}}{\frac{\Delta C}{L} Area} \quad (5.2)$$

Crossover frequencies and the trends observed theoretically for the total mass transfer also remain the same for D_{eff} . This is how the experimental and theoretical values will now be compared.

Table 5.3 Total error in the theoretical values for the effective diffusion coefficient

Variable	Error in Variable	Deviation in Effective Diffusion Coefficient At 1 Hz (2 π rad/sec)
Amplitude (A)	+ / - 0.8 %	+ 1.6 %, - 1.5 %
Diffusion Coefficient (D)	+ / - 1.0 %	+ 1.4 %, - 0.9 %
Kinematic Viscosity (ν)	+ / - 0.7 %	+ 0.2 %, - 0.08 %
Outer Radius (R_{out})	+ / - 1.3 %	+ 1.0 %, - 1.2 %
Total Error		+ 3.1 %, - 3.0 %

For the experimental data retrieved, each data point is affected by the accuracy of the concentration measurement and the accuracy of the flow rate from the mass flow controllers. The error associated with the data from the experiments is given in Table 5.4.

Table 5.4 Total error in experimental values for the effective diffusion coefficient

Variable	Error in Variable	Deviation in the Effective Diffusion Coefficient
Flow Rate	+/- 8.9 %	+/- 5.8 %
Reservoir 1 Outlet Concentration (C_1)	+/- 4.0 %	+/- 4.0 %
Reservoir 2 Outlet Concentration (C_2)	+/- 4.0 %	+/- 4.2 %
Outer Radius (R_{out})	+/- 1.3 %	+/- 2.6 %
Inner Radius (R_{in})	+/- 1.6 %	+/- 5.8 %
Total Error (open tube)		+/- 11.7 %
Total Error (annulus)		+/- 17.5 %

5.4 Experimental Results for an Open Tube

With the sensitivity analysis complete, the results for various experiments for the open tube and later for the annular configuration will now be presented along with the models that theoretically predict the phenomena observed. These experiments, along with further discussion on the experimental and theoretical results, complete the analysis on the mass transfer, or effective diffusion coefficient, in different configurations and the separation of dilute species. Unless otherwise noted, the normal operating parameters for the experiments are given in Table 5.5.

Table 5.5 Normal operating parameters for experiments

Parameter	Value
Peak-to-Peak Amplitude (A)	15 cm
Tube Length (L)	134 cm
Outer Tube Radius (R and R_{out}) (Open tube and Annulus)	0.80 cm
Inner Tube Radius (R_{in}) (Annulus only)	0.635 cm
Flow Into Both Reservoirs	900 sccm

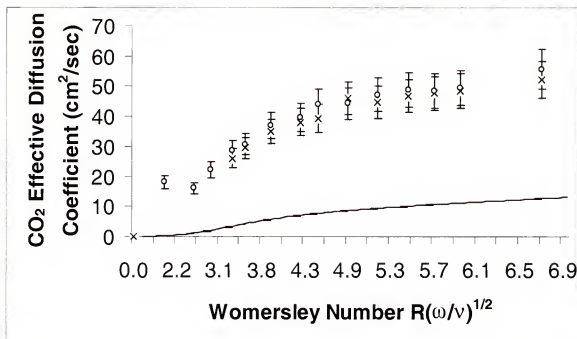


Figure 5.2 Experimental results for the effective diffusion coefficient of carbon dioxide versus Womersley Number for two separate runs. The theoretical curve is at the bottom of the figure.

When an experiment for an open tube possessing dilute amounts of CO₂ and CH₄ in N₂ was performed, Fig. 5.2 shows the results for the effective diffusion coefficient of CO₂ for various frequencies of oscillation. The curve at the bottom of the figure represents the theoretical prediction over the frequency range. The experiment was run twice, and there was a reasonable reproducibility between the results; however, there is not good agreement between the experiments and the theory for the value of the effective diffusion coefficient. The general trend of increasing effective diffusivity as frequency increases is apparent, but the experimental values for the effective diffusion coefficient are approximately an order of magnitude *higher* than the theoretical predictions. The discrepancy will be discussed later in this chapter. This is also evident in Fig. 5.3 that depicts the effective diffusion coefficient of CH₄ for various frequencies. However,

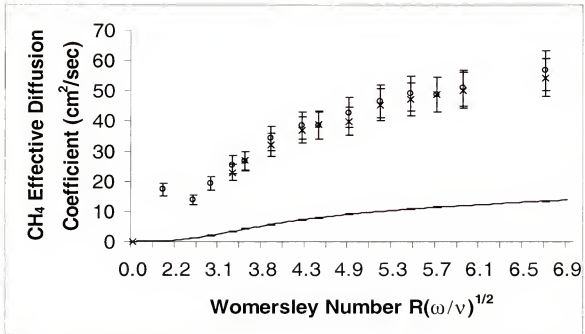


Figure 5.3 Experimental results for the effective diffusion coefficient of methane versus Womersley Number for two separate runs. The theoretical curve is at the bottom of the figure.

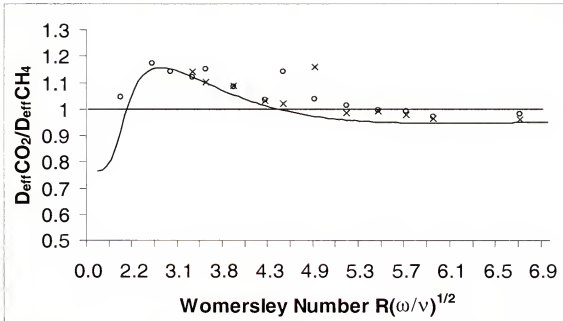


Figure 5.4 Experimental separation ratios for methane and carbon dioxide versus Womersley number compared to the theoretical model.

when the ratio of the effective diffusivities is plotted versus frequency in Fig. 5.4, the experimental and theoretical results show an excellent agreement. A maximum in the experimental result is clearly seen along with the *second* crossover where the fast diffusing CH_4 regains the advantage over the slower diffusing CO_2 . Both the maximum and the crossover occur at frequencies close to the predicted value. This is encouraging because the physics of periodic flow predicted by the calculation is also seen in the experiments even though the absolute values of the effective diffusion coefficient are not in agreement.

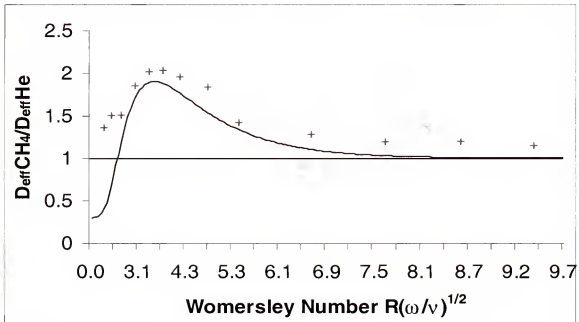


Figure 5.5 Separation ratio for helium and methane showing both the experiments and theoretical model.

This is also evident in Fig. 5.5 that shows the ratio of the mass transfer for a helium and CH_4 system. The general shape of the curve is clearly observed with the maximum in the ratio occurring at nearly the frequency predicted. The values are consistently higher than the model, however, and the values at low frequencies (~ 0.1 Hz

or 0.6 rad/sec) do not follow the trend. The lower frequencies were difficult to measure experimentally because the inertia of the flywheel interfered with achieving a constant angular speed as the rotation became very slow. Furthermore, some sensitivity is lost in the concentration measurement since Argon was used as the carrier gas in the gas chromatograph in order to measure the Helium.

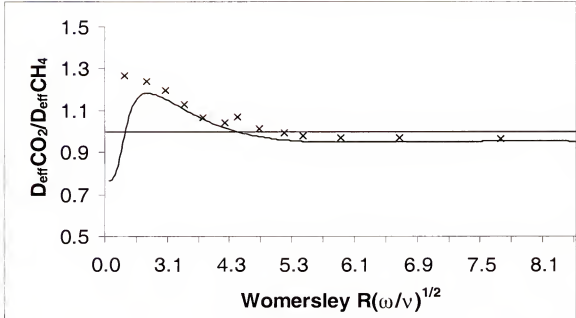


Figure 5.6 Separation ratio for methane and carbon dioxide versus Womersley number for an increased peak-to-peak amplitude of 20 cm.

Finally, the peak-to-peak amplitude for the $\text{CH}_4 - \text{CO}_2$ system was increased to 20 cm. The ratio of the effective diffusion coefficients for this system is shown in Fig. 5.6. Again, the general trend of the separation ratio is still present with deviations occurring at small Womersley numbers as before.

The physics of the model are clearly evident in the separation ratios produced by the $\text{CH}_4 - \text{CO}_2$ for both amplitudes and the $\text{He} - \text{CH}_4$ systems. As the absolute values of the effective diffusion coefficient are still very much higher than those predicted for these

systems, some other factors not accounted for in the experiments must be causing the disparity. This will be discussed in the subsequent section.

5.5 Discussion of Results for the Open Tube

Explanations are now provided to give possible reasons for the disparity between some of the experimental results and the models. These conjectures are based on physical arguments and on a few small experiments to analyze the effect of different factors.

For the CH_4 - CO_2 system, the absolute difference between the experimental effective diffusion coefficient and the predicted value is given in Fig. 5.7. Upon observation, it is seen in Fig. 5.7 that the difference in values becomes progressively greater for increasing frequency. This indicates that the theoretical values are not simply shifted lower due to a common factor not included in the model. In other words, adding a constant to the predicted values will not make them coincide with the experimental results. Further, the difference is also greater for a larger amplitude of oscillation although the difference is the same for both dilute species at both amplitudes. Whatever is causing the disparity affects both species in a similar way. Finally, the *percent* difference between the experimental and theoretical values is shown in Fig. 5.8. The percent difference is large for both amplitudes at small Womersley numbers and slowly becomes better as the Womersley number increases. This shows that on average, the disparity between results for both amplitudes is roughly the same.

On first observation, the difference between the measured effective diffusion coefficient for each species and that calculated seems to be related to an overall steady

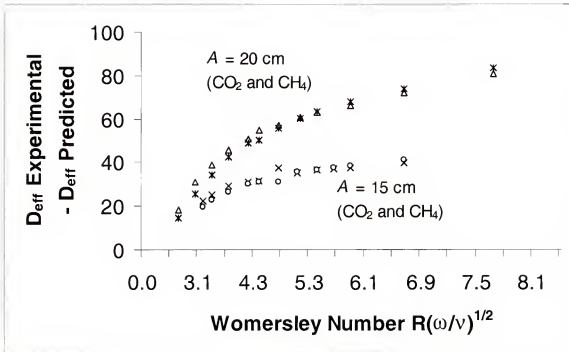


Figure 5.7 Difference between experimental results and predicted values for the methane and carbon dioxide systems for the two peak-to-peak amplitudes used.

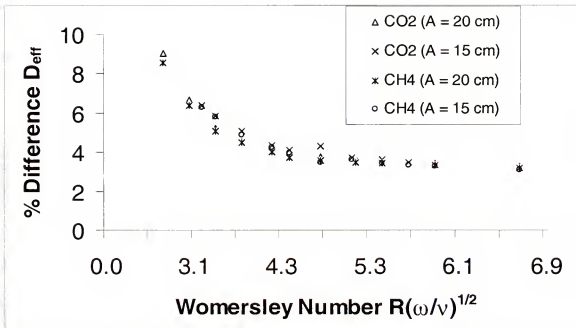


Figure 5.8 Percent difference between experimental results and predicted values versus Womersley number for methane and carbon dioxide at the two amplitudes used.

flow in the direction of the concentration gradient superimposed on the oscillations. If a steady flow were present, the mass transport of each species would increase because a net flow exists between the reservoirs. Although careful consideration has been given to ensure that the flow rates into and out of each reservoir are within the experimental error of the instruments, there could still be a pressure drop along the tube from Reservoir 1 to Reservoir 2. Calculations have shown that only a small pressure drop is needed to dominate the mass transport in periodic flow.¹

An experiment was then performed to measure the significance of the steady flow, if it were present, that would occur during the runs. The same experimental procedure discussed previously was followed except the motors were not started. Concentration measurements were made and a measurable amount of both CO₂ and CH₄ were detected in the Reservoir 2. The motors were then turned on and run at different Womersley numbers with a 20 cm amplitude. The effective diffusion coefficient was then recorded and compared to the theoretical value knowing the amount contributed by the steady flow. These comparisons are noted in Table 5.5. As can be seen, the steady flow does contribute to the overall effective diffusion coefficient, but it does not totally account for the disparity.

¹ This calculation was done by an undergraduate student in our group and is given in Appendix E. A pressure drop of 2.3 g/(cm*sec²) will give the same effective diffusion coefficient for CO₂ in an open tube as an oscillating system operating at 3.14 rad/sec ($W = 3.8$) with a peak-to-peak amplitude of 15 cm.

Table 5.5 The effect of steady flow on the effective diffusion coefficient

Species	Effective Diffusion Coefficient for Steady Flow	Overall Effective Diffusion Coefficient	Effective Diffusion Coefficient Minus Steady Flow	Theoretical Prediction
CO ₂ / CH ₄ (W = 0)	16 / 14 (cm ² /sec)	16 / 14 (cm ² /sec)	16 / 14 (cm ² /sec)	0.141 / 0.184 (cm ² /sec)
CO ₂ / CH ₄ (W = 3.6)	16 / 14 (cm ² /sec)	60 / 55 (cm ² /sec)	44 / 41 (cm ² /sec)	10 / 9 (cm ² /sec)
CO ₂ / CH ₄ (W = 5.2)	16 / 14 (cm ² /sec)	83 / 83 (cm ² /sec)	67 / 69 (cm ² /sec)	18 / 19 (cm ² /sec)
CO ₂ / CH ₄ (W = 6.3)	16 / 14 (cm ² /sec)	97 / 100 (cm ² /sec)	81 / 86 (cm ² /sec)	22 / 23 (cm ² /sec)

It would make sense that the steady flow would *not* entirely justify the disparity as the difference plotted in Fig. 5.7 shows that it steadily increases. The steady flow may be the reason for the discrepancy of the effective diffusion coefficient at *small* Womersley numbers, and it does explain the difference in the values for the separation ratio seen in Fig. 5.6 at low Womersley numbers. Although the faster diffusing CH₄ is expected to have a higher diffusion coefficient at small Womersley numbers, the steady flow prevents this from occurring.

It was also reasoned that in addition to a steady flow occurring in the experiments, a secondary flow caused by pressure fluctuations induced by the tube motion could alter the results. Because the reservoirs are of finite volume, the ends of the tubes could displace some of the volume in the reservoir and cause it to be redirected back down the tube. At higher frequencies (1.5 Hz or greater), the reading on the MFC's for the outlets oscillate +/- 2 sccm suggesting that the pressure in the reservoirs oscillates as well. For this experimental apparatus, Poplasky [1] calculated the pressure change in the reservoir due to the tube displacement as well as the displacement caused by fluid adhering near the vicinity of the tube surfaces being brought into and out of the reservoir during

oscillation. He calculated that the resulting pressure fluctuation for one tube would be about 0.7 % and would not appreciably affect the experiments. However, Rice and Eagleton [18] also considered this problem by calculating the total mass transfer for a configuration where the finite thickness of the oscillating outer tube causes periodic pressure fluctuations. Rice and Eagleton conjecture that a tube oscillating sinusoidally will cause an appreciable periodic pressure drop 180° out phase from the normal flow profile, and the amount of the pressure drop depends on the thickness of the outer tube. Their calculation also assumes that the volume displacement is redirected down the tube instantaneously so that the pressure in the reservoir is kept constant. This implicitly assumes that the fluid is incompressible, but their correction will still be analyzed here to determine its effect on the mass transfer and separation. Rice and Eagleton show that as the ratio of the tube annular cross sectional area to the flow cross-sectional area increases, the total mass transfer also increases.

Rice and Eagleton's model for the boundary driven total mass transfer with induced pressure oscillations is given in Equation 34 of their study [18]. This equation was then used to find the effective diffusion coefficient for my experimental configuration. The outer radius of the open tube for these experiments is 0.95 cm giving an outer tube area to flow area ratio of 0.41. The results of this theoretical model are shown in comparison to the previous theoretical model (no pressure oscillations) and to the experiments in Fig. 5.9. As can be seen, the induced pressure oscillations greatly enhance the overall effective diffusion coefficient. Although the model is still lower than the experimental value, it is certainly a step in the right direction. The separation ratio was also calculated for the Rice and Eagleton model and given in Fig. 5.10 for the CO_2 –

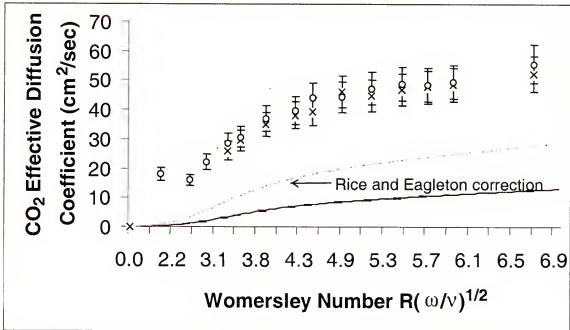


Figure 5.9 Effective diffusion of carbon dioxide versus Womersley number shown again including the correction by Rice and Eagleton.

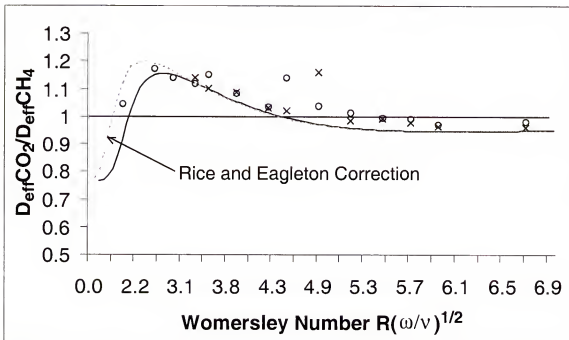


Figure 5.10 Separation ratio the methane and carbon dioxide system versus Womersley number including the correction by Rice and Eagleton.

CH₄ system. The separation ratio is altered slightly, especially at low Womersley numbers, but it is still in reasonable agreement with the experimental results. At large Womersley numbers, it appears that the pressure oscillations have little effect on the separation ratio as both curves asymptote towards each other. The separation ratio of this model was also compared to the other configurations as seen in Fig. 5.11 for the CH₄ – He system and in Fig. 5.12 for the CO₂ – CH₄ system operating at a higher amplitude. The model shows a greater deviation for the CH₄ – He system in terms of the separation ratio, but there is still reasonable agreement with the experiments. For the CO₂ – CH₄ system operating at an peak-to-peak amplitude of 20 cm, the new model also closely predicts the experimental results. It is interesting to note that not only do the induced pressure oscillations increase the mass transfer of each species, it also increases the separation as seen in these figures. From the results already found, it is indeed true that

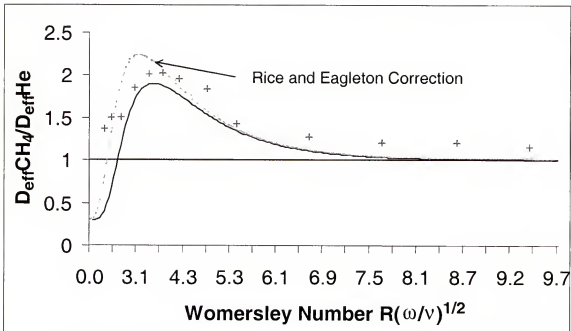


Figure 5.11 Separation ratio for the helium and methane system versus Womersley number including the correction by Rice and Eagleton.

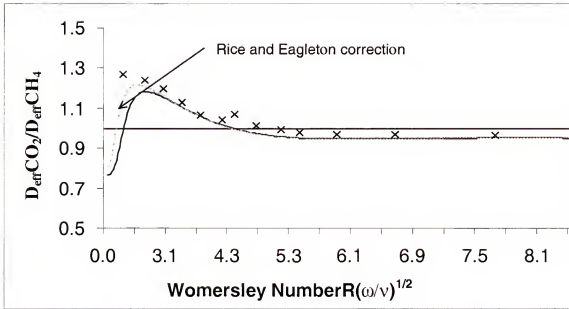


Figure 5.12 Separation ratio for the methane and carbon dioxide system at a peak-to-peak amplitude of 20 cm. The Rice and Eagleton correction is included.

more consideration needs to be given to the fluid mechanics that occur in the experiments.

Another area of consideration is in the validity of the assumption that the end effects can be ignored during tube motion. U. H. Kurzweg indicated that during some of his experiments with pressure driven oscillations he noticed a turbulent region on the order of the one-half the oscillation amplitude present at both ends of the tubes. Now this is something only observed and not accurately measured, but it could mean significant mixing is present at the ends of the tubes. This would affect the concentration gradient $(c_1 - c_2) / L$ because if mixing occurs on the order of A , then the effective length would then be $L - A$. This mixing would then coincide with the larger amplitude system having a greater difference in the effective diffusion coefficient values (in addition to the effect of induced pressure oscillations). However, upon only making this correction, it does not

totally make up the difference in the experimental and predicted values. It does, however, continue to take another step in the right direction, and since this observation has not been measured, turbulent mixing at the ends may affect the mass transfer more than reasoned here and should be investigated further.

Another possibility is that secondary flows may be occurring in the tube in a manner other than that predicted by Rice and Eagleton and Poplasky. These secondary flows must not be radically changing the shape of the velocity profiles since the physics is retained experimentally, but secondary flows may be enhancing the profiles in the direction of the concentration gradient in some way. It seems that an extra pumping mechanism is causing both species to move in the direction of the concentration gradient in an enhanced way. It was reasoned that perhaps an imbalance in the symmetry of the

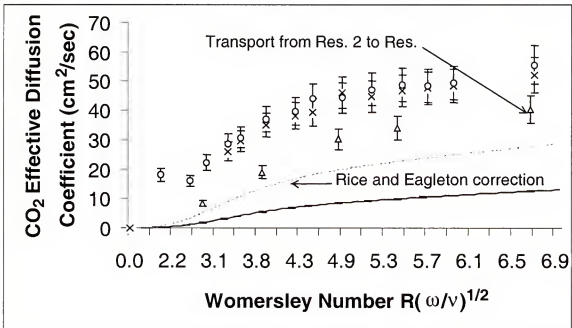


Figure 5.13 Effective diffusion coefficient for carbon dioxide versus Womersely number including the correction by Rice and Eagleton and experimental results for transport from Reservoir 2 to Reservoir 1.

reservoirs causes a flow profile to favor transport from Reservoir 1 to Reservoir 2. An experiment was performed to see what would happen if the direction of mass transfer was switched to transport from Reservoir 2 to Reservoir 1. These results for the effective diffusion coefficient from Reservoir 2 to Reservoir 1 have been added the previous data and are shown in Fig. 5.13 along with the previous experimental results and both theoretical models. The results for CO_2 are a little lower than the earlier effective diffusion data, but this may be due to two reasons. First, there may be counterflow from Reservoir 1 to Reservoir 2 that would hinder the effective diffusion coefficient for species diffusing against this flow. Second, there may very well be an imbalance in the symmetry that favors transport from Reservoir 1 to Reservoir 2; however, the imbalance is not enough to account entirely for the discrepancy in the experimental and theoretical results as the experimental results are still higher. It does come closer to the theoretical values predicted by Rice and Eagleton as seen in the figure.

There appear to be many factors that affect the mass transfer of the system. Steady flows, pressure induced oscillations, observed end effects, and an imbalance in symmetry are the only imperfections in the flow that have been reasoned thus far. An extra pumping mechanism may still be present, but it may arise from other means such as complicated flows occurring at the ends of the tubes that effect the profiles throughout the length of the tubes. In order to fully understand what is happening, I believe that a full numerical calculation needs to be done on the flow that occurs in the reservoirs, at the ends of the tubes, and all along the tube length. Of course in annular flow, it is reasonable to assume the flow will be affected even more because of the motion of two tubes. The results from experiments in periodic annular flow will be analyzed next.

5.6 Experimental Results for an Annular Geometry

Results for the effective diffusion and separation in periodic annular flow will now be presented. This will include experiments for tubes oscillating in phase, tubes oscillating out of phase, and tubes oscillating in phase at different amplitudes. The experimental results will be compared to theoretical trends to see if the models are indeed valid.

First, the effective diffusion coefficient for a $\text{CO}_2 - \text{CH}_4$ binary system present in a nitrogen carrier for tubes oscillating *in phase* is given in Fig. 5.14. The radius of the outer tube remains 0.8 cm as it was for the open tube geometry, but the added inner tube has an outer radius of 0.635 cm. Therefore, the flow area of the annular region for these experiments is much smaller than the flow area for the open tube experiments analyzed

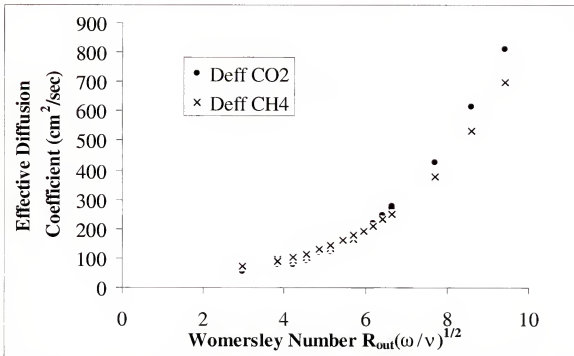


Figure 5.14 Effective diffusion coefficients for a methane and carbon dioxide system versus Womersley number for the annular configuration.

previously. In Fig. 5.14, the first crossover is clearly seen as the faster diffusing CH_4 initially has the higher effective diffusion coefficient at small Womersley number, but the slower diffusing CO_2 overtakes methane and has a higher effective diffusion coefficient for larger Womersley numbers. The crossover frequency for this configuration is at a Womersley number of approximately 5.7. As the overall area provided for the mass transfer is smaller for the annular experiments than for the open tube, this accounts for the first crossover to occur at a higher Womersley number for an annular configuration. A second crossover would not occur until a much higher Womersley number, but it may not be possible to experimentally see this crossover with the existing experimental setup. This crossover Womersley number would require a frequency that the experimental assembly may not be able to handle, or possibly at a frequency not in the laminar regime.² A reasonable alternative to find the second crossover at a lower frequency would be to use larger outer tubes to create a greater cross-sectional area. This could be an experiment done by future workers on this apparatus.

The experimental predictions for the effective diffusion coefficient will now be compared to theoretical predictions. As has been mentioned before, Maple® has difficulty in evaluating expressions with complicated forms of the Bessel functions. When trying to use Maple® to give values for the parameters of this experiment, it was unable to give an accurate analysis over the experimental range of Womersley numbers. Therefore, the Maple® program devised by Poplasky for periodic annular flow for tubes

² According to Kurzweg et al. [27] on the onset of turbulence in oscillating pipe flow, a frequency of 300 rad/sec (48 Hz) would be at the limiting frequency for laminar flow in an open tube with an amplitude of 15cm, radius of 0.8 cm, and a kinematic viscosity of $0.136 \text{ cm}^2/\text{sec}$. If the characteristic length for annular periodic flow were taken to be

oscillating in phase will be employed. In his program, the model was calculated using a FORTRAN code imbedded in the Maple® worksheet. This FORTRAN code evaluated the Bessel functions involved in the model using the series expansion of the Bessel functions up to a large number of terms. Although Poplasky's results have not been verified, they do make predictions that coincide with results expected on physical grounds. Poplasky's model and experimental results for the effective diffusion coefficient of CO_2 are shown in Fig. 5.15. The experimental values are much higher than those predicted in the theoretical model. In fact, the experimental effective diffusion coefficient is much higher than that given for an open tube. This goes against what was seen in Chapter 3 comparing the two geometrical configurations. However, this does follow what would be expected given that two tubes are now greatly altering the flow.

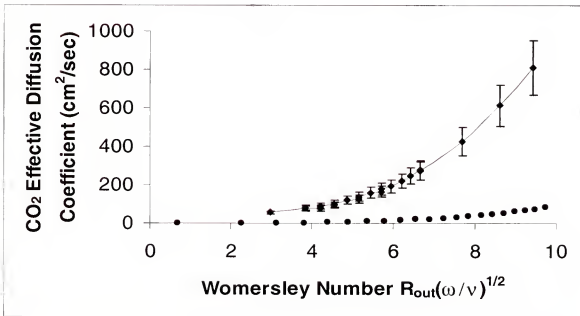


Figure 5.15 Experimental effective diffusion coefficient of carbon dioxide (upper points) compared to the theoretical prediction using Poplasky's program (lower points).

$R_{out} - R_{in}$, 300 rad/sec would still be the limiting frequency for laminar periodic flow using the same model as the open tube.

Again, according to Rice and Eagleton for an open tube, the larger the ratio of tube area to the flow area, the larger the total mass transfer. For my annular configuration, the flow area has become smaller, and an added pressure induced oscillation is caused by the solid inner tube. It would indeed follow that the effective diffusion coefficient would increase greatly for my annular configuration, even more so than that for the open tube. A model should be developed similar to that of Rice and Eagleton for periodic annular flow to analyze the effect of pressure induced oscillations for this annular configuration.

In looking at the separation ratio for the $\text{CO}_2 - \text{CH}_4$ system depicted in Fig. 5.16, it is clearly seen that the experimental results are dramatically different than the model. Although they appear to agree at larger Womersley numbers, it is unclear whether they will continue to do so as the Womersley number increases even more. This suggests that

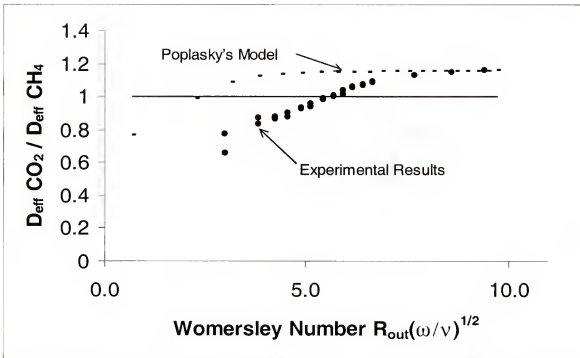


Figure 5.16 Separation ratio for methane and carbon dioxide versus Womersley number with Poplasky's model and the experimental results.

the flow profiles have been significantly altered in some way not accounted for in the model. As with the open tube, these alterations could include pressure induced oscillations, end effects, or some other mechanism not yet considered. As with the open tube, a full numerical computation should be performed to model the flow that occurs in a periodic annular configuration with walls of finite thickness and finite reservoir volumes.

Moving on to different configurations associated with annular periodic flow, experiments were also done on tubes oscillating 180° out of phase. Experiments were performed over a typical range of frequencies: $\omega = 1.9 - 12.6$ rad/sec or Womersley numbers based on the outer tube radius of $2.8 - 7.3$. It was not until an oscillation frequency of about 9.4 rad/sec ($W = 6.3$), however, that a detectable amount of species was present in Reservoir 2. Table 5.6 shows the effective diffusion coefficient for CO_2 and CH_4 for tubes oscillating 180° out of phase compared to the values for tubes oscillating in phase from experiments conducted the following week.

Table 5.6 Effective diffusion coefficients for tubes oscillating in phase and 180° out of phase.

Womersley Number $R_{\text{out}}(\omega/\nu)^{1/2}$	Out of phase D_{eff} CO_2 (cm^2/sec)	In phase D_{eff} CO_2 (cm^2/sec)	Out of phase D_{eff} CH_4 (cm^2/sec)	In phase D_{eff} CH_4 (cm^2/sec)
2.8		37		57
5.2		120		121
5.7		158		153
5.9		174		164
6.1		189		176
6.3	27		19	
7.3	40		32	

As can be seen, the effective diffusion coefficient for both species is much less for the tubes oscillating out of phase than for the tubes oscillating in phase. According to the

theory outlined in Chapter 4, there is indeed an intermediate range of Womersley numbers where the effective diffusion coefficient is expected to be less for out of phase tubes as seen in these experiments. However, the theory also showed that at small Womersley numbers the effective diffusion coefficient should be greater for tubes out of phase and large Womersley numbers indicated no difference between either configuration. Now, it may still be true that at large Womersley numbers both the in phase and out of phase configurations may experimentally be the same, but this may occur at frequencies larger than the apparatus is able to accommodate. At small Womersley numbers, the enhancement of the effective diffusion coefficient is not seen experimentally. This may be due to complicated flow fields that have been hampering our previous experiments. Indeed, if pressure induced oscillations were introduced in the model, tubes oscillating out of phase would produce a complex flow field. Further, the predictions from the flat plate geometry of Chapter 4 may be insufficient in predicting the trends of this experimental setup.

Finally, experiments were conducted for tubes oscillating *in phase* but at different amplitudes. First, the inner tube's peak-to-peak amplitude was increased to 20 cm while the outer tube's peak-to-peak amplitude was held at 15 cm. Measurements were taken over a reasonable range of frequencies: 3.14 – 12.6 rad/sec. Then, the peak-to-peak amplitude of the inner tube was decreased to 15 cm while the outer tube was increased to 20 cm to see if there was a difference in which tube was oscillating faster. Measurements were again taken over the same range of frequencies. The results of both experiments are shown in Fig. 5.17 for the effective diffusion coefficient of CO₂. The results for the most recent experiments for both tubes oscillating at 15 cm are also provided for reference.

There is a marked increase in the effective diffusion coefficient for tubes oscillating at different amplitudes over tubes oscillating at the same amplitude. This agrees with the phenomena predicted for flat plates oscillating at different amplitudes. It also appears that for larger Womersley numbers the effective diffusion coefficient is slightly greater for the outer tube oscillating at the higher frequency. This is somewhat of a surprise as one might expect the inner tube to produce a greater pressure induced oscillation because of its greater cross-sectional area; therefore, it would provide a greater effective diffusion coefficient if it oscillated at the larger amplitude. It may be that mass transfer favors the outer tube oscillating faster because of the asymmetry in the field in an annular region. Of course, because of experimental error, the true result may be that both configurations

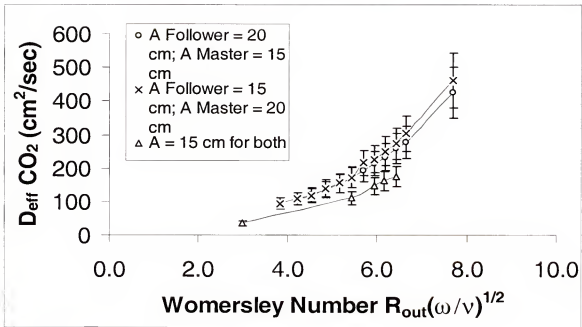


Figure 5.17 Experimental results for the effective diffusion coefficient of carbon dioxide versus Womersley number for tubes oscillating at different amplitudes as given in the legend. Master is the outer tube and Follower is the inner tube.

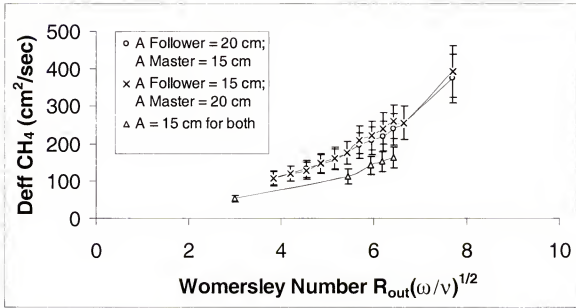


Figure 5.18 Experimental results for the effective diffusion coefficient of methane versus Womersley number for tubes oscillating at different amplitudes as given in the legend. Master is the outer tube and Follower is the inner tube.

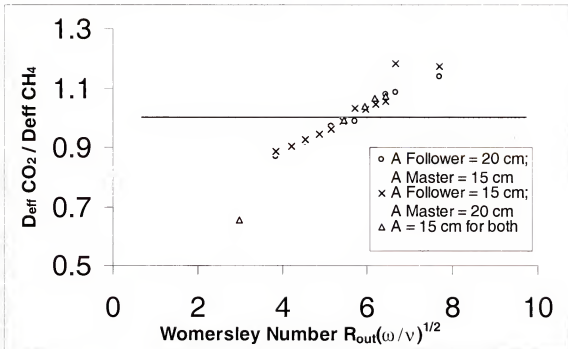


Figure 5.19 Separation ratio for tubes oscillating at different amplitudes. Master is the outer tube and Follower is the inner tube.

give the same values or that the faster inner tube has the greater effective diffusion.

These results are further verified in Fig. 5.18 depicting the effective diffusion coefficient of methane.

For the separation ratio, it is seen that there is little difference between which tube oscillates at the larger amplitude. Further, there appears to be little difference between oscillating tubes at different amplitudes and oscillating tubes at the same amplitude. These results are shown in Fig. 5.19. From the theory provided by the simple flat plates model, one would have expected the tubes oscillating at the different amplitudes (one being a larger amplitude) would give a greater separation. This may be true at even higher Womersley numbers, or the secondary flows that have been a problem have added new phenomena not considered in the model.

It is clear that the present experimental apparatus produces results that have not been accounted for in the previous theoretical models. For the open tube, some added considerations such as steady flow, induced pressure oscillations, and turbulent mixing in the ends seem to be taking the proper steps in the right direction to predict experimental results. However, for the annular configuration, it is clear that the models are not sufficient to predict experimental findings. Either the experimental apparatus needs to be modified in order to make the flows in the reservoirs and the tubes more simplistic, or the theoretical models need to be changed to describe the complicate flows existing in the present apparatus.

5.7 Endnote 1: Verification That the Data for the Annular Geometry is Statistically Different

Two series of experiments, one made with CO_2 in N_2 and the second made with CH_4 in N_2 each show a curvilinear relationship between D_{eff} and Womersley number as

seen in Fig. 5.14. In order to prove that the two relationships have a crossover, i.e. a value of Womersley number for which $D_{eff}CO_2 = D_{eff}CH_4$, both sets of data were first fit to exponential curves (which may be valid for small Womersley numbers). For CO_2 ,

$$D_{eff} = 15.1 \exp(0.44W) \quad r = 0.994 \quad (5.3)$$

for 22 data points, and for CH_4 ,

$$D_{eff} = 22.6 \exp(0.37W) \quad r = 0.993 \quad (5.4)$$

for 22 data points. The high correlation coefficient, r , is an indication of a successful fit to the data. The experimental data and trend lines are depicted in Figures 5.20 and 5.21.

The curve fit was then written in a linear way as

$$\ln(D_{eff} CO_2) = \ln(15.1) + 0.44W \quad (5.5)$$

and

$$\ln(D_{eff} CH_4) = \ln(22.6) + 0.37W \quad (5.6)$$

In order for a crossover to exist at a Womersley number of approximately 5.8, the slopes of these two curves must be significantly different. An analysis of variance was performed on the relationship between the Equations 5.5 and 5.6 and their respective data points. Then, a t – test was made on the slopes of the two curves, and the results showed that the slopes were significantly different ($t = 4.33$; $P < 0.006$ for 40 degrees of freedom). This proves that a crossover does indeed exist and that it is unlikely due to the randomness of the data.

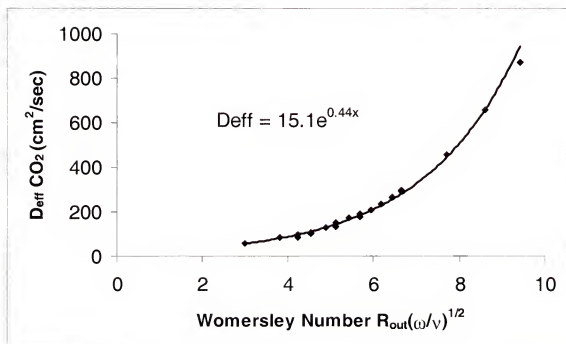


Figure 5.20 Experimental data for the effective diffusion coefficient of carbon dioxide in an annular geometry with added trend line.

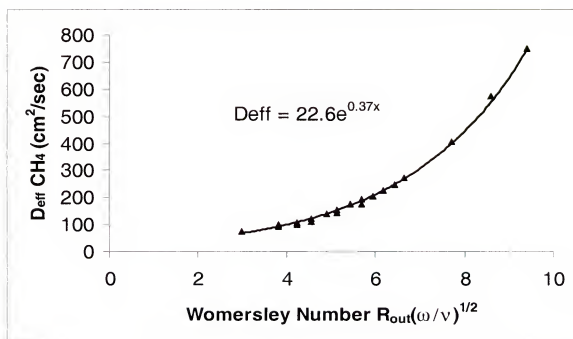


Figure 5.21 Experimental data for the effective diffusion coefficient of carbon dioxide in an annular geometry with added trend line.

CHAPTER 6 CONCLUSIONS AND FUTURE IDEAS

6.1 Conclusions on This Work

Periodic flow has shown to greatly enhance the mass transport of a dilute species present in a carrier over that due to pure molecular diffusion. Although there is no net flow of the carrier fluid, a net transport of species exists as radial concentration gradients generated from the flow profile allow a dilute species to diffuse from the core to the boundary and back again taking advantage of the convection. If two dilute species were present in a carrier, then a separation is possible as both species diffuse at different rates. This phenomenon was observed theoretically for different geometries and further verified by experimental results.

For an oscillating flat plate below a fluid of infinite extent, it was found that the total mass transfer varies linearly with an increase in the frequency of oscillation. For a CO_2 -He binary system present in a nitrogen carrier, the slower diffusing carbon dioxide initially had the lower mass transfer, but as frequency increased, the mass transfer for carbon dioxide was greater than that from the faster diffusing helium. This calculation required an effective transverse length over which 99.9 % of the mass transfer occurred. This effective length changed with increasing frequency for gases, so this motivated further analysis in the area of finite geometries.

The first finite geometry studied was periodic flow between two flat plates. Now, periodic flow can be generated by imposing a periodic pressure drop such as from an

oscillating piston or by oscillating both plates in phase. It was shown that these two configurations are physically the same, and in looking at the convective mass transfer per power supplied to both the pressure driven and boundary driven systems, they are quantitatively exactly the same. With this result known, the mass transfer for three binary systems (He – X, He – CO₂, and He – CH₄, where species X is an unnamed species with a specified diffusion coefficient) was analyzed in periodic pressure driven flow in a flat plate geometry.

The results for these three binary systems showed that crossover frequencies exist where the mass transfer of each species is the same. For the He – X system, only one crossover was present. For He – CO₂, two crossovers existed, and three crossovers were evident for the He – CH₄ binary system. These results demonstrate that the faster diffusing species in a system may or may not possess the higher mass transfer in periodic flow. The difference in diffusion coefficients, the kinematic viscosity, the frequency of oscillation, and the channel width all contribute to the total mass transfer of the system. While, at very low frequencies, the faster diffusing species will transport at a higher rate than the slower diffusing species, at higher frequencies, the slower diffusing species has a higher mass transfer because the faster diffuser resides near the slow moving boundary of the system. At even higher frequencies, neither species has enough time to get to the slow moving boundary in a cycle, so the advantage is regained by the faster diffusing species. At very large frequencies, reverse flow in the velocity profile hinders the faster diffusing species as it moves into the regions of reverse flow more than the slower diffusing species giving the slow diffuser the advantage again in the total mass transfer. In this way, the frequency plays a role in the dominance of the mass transfer of one

species over another. Kinematic viscosity also plays an important role as a larger kinematic viscosity delays the frequency at which “wiggles” and reverse flows occur. The total mass transfer is therefore dependent on how each system property affects the movement of the particles of each species in taking advantage of the convection produced by the periodic flow. It is the interaction between the system properties and their collective effect on the total mass transfer that is conveyed in the section on periodic flow in a channel.

Periodic flows in an open tube and in an annular region were two circular geometries also studied in this thesis. For an open tube and annulus with the same cross-sectional flow area, the open tube exhibits a higher transport of a dilute species than the annular geometry. In terms of separation, the annulus provided a slightly higher separation of two dilute species than the open tube. Not only was the separation greater, the frequency where the maximum separation could be achieved for the annulus also gave a higher transport of both dilute species than the maximum for the open tube. These results demonstrated that periodic flow in an annular region is a viable option to achieve a reasonable separation at a high throughput (mass transfer).

In further studies on periodic annular flow, imperfections associated with this configuration were examined. One of these imperfections included an eccentric geometry where the inner tube of the annulus is slightly off center from the outer tube. Calculations on the eccentric geometry showed that the total mass transfer was less than that for two concentric tubes; however, the separation of two dilute species increased for an eccentric annulus. Further considerations in periodic annular flow included tubes oscillating out of phase and tubes oscillating in phase at different amplitudes. Simple

calculations using periodic flows between two flat plates were used to model the general trends that these configurations would have on an annular geometry. For plates oscillating out of phase, it was shown that the mass transfer initially increased, then decreased, and then eventually matched that for plates oscillating in phase as the frequency increased. It was also shown that the separation radically changed for plates oscillating out of phase. For plates oscillating in phase, the mass transfer was greater if one of the plates oscillated at a larger amplitude and lower if one of the plates oscillated at a lower amplitude. The separation was also greater for the larger amplitude configuration and lower for the lower amplitude configuration. This came as no surprise.

Experiments demonstrated that some of the models developed for the open tube and annular geometries were somewhat deficient. The effective diffusion coefficient predicted from the theoretical models was much less than what was seen in experimental runs. However, the calculated separation ratio closely agreed with the separation ratio achieved in experiments. This is in indication that the physics behind periodic flow of binary species is well approximated. Further considerations such as a steady flow from one reservoir to the other, mixing at the ends, imbalances in experimental symmetry, and pressure induced oscillations were analyzed. The most promising results for the open tube were the pressure induced oscillations model developed by Rice and Eagleton [18]. This model increased the total mass transfer bringing theory and experiments to closer agreement without significantly altering the separation ratio.

For periodic flow in an annular region, both the mass transfer and the separation of dilute species greatly differed between projected results and experimental values. However, a crossover frequency was still observed in the experiments, which was

encouraging. All of the aspects that caused the deviations for the open tube experiments also apply to annular periodic flow, especially the pressure induced oscillations. With the addition of the inner tube and a decrease in the cross-sectional flow area for an annulus, pressure induced oscillations would alter the results more for the annular geometry than for the open tube. It is these types of secondary flows that may account for deviations observed experimentally not considered in the models.

The main contributions of this work to the field of oscillating flows is as follows:

- The physics of the interaction between dilute species and the oscillating fluid were carefully explained. In particular, the ability of a species to remain near the fast moving flow regions and its dependence on the thermophysical parameters of the system were presented.
- The relationship between a flow induced by an oscillating boundary and by and oscillating pressure drop was also provided.
- The approximation that the concentration can be written as the sum of an axial gradient and radial and time dependent term was mathematically proven to be an appropriate approximation using the method of moments.
- The effect of geometry in oscillating flows on the mass transfer and separation of species was shown for an open tube and an annular configuration. Further, an eccentric geometry in periodic annular flow had a negative effect on the mass transfer but increased the separation.
- Configuration where tubes oscillate out of phase and tubes oscillating at different frequencies demonstrated that these are viable options in increasing the mass transfer and/or separation depending on the parameters of the system.

- Experiments were performed that showed close agreement with the models in terms of separation, but improvements need to be made to predict the overall mass transfer.

6.2 Future Consideration for Improvement on This Work

Improvements need to be made in order to improve the comparisons between the theoretical model and experimental results. This can be accomplished in two different ways:

1. Add to the theoretical models to include secondary flow considerations
2. Modify the experimental setup to minimize the secondary flows that significantly alter the flow profiles.

Some considerations that can be added to the theoretical models include:

1. Adjust the pressure induced oscillation of Rice and Eagleton to account for the compressibility of the gas in the reservoirs.
2. Pressure induced oscillations for the annular configurations using the ideas of Rice and Eagleton.
3. Including a steady flow term to account for a net flow that exists either with or against the concentration gradient.
4. Do a complete analysis on the complex flows that exist in the tubes, at the tube ends, and in the reservoirs. This would most likely require a numerical calculation using software such as FLUENT® or some other numerical procedure.

Modifications on the existing experimental setup may include:

1. Make the tube length larger and operating amplitudes smaller to limit the turbulent mixing that may exist at the ends of the tubes.

2. Construct larger reservoirs. Pressure induced oscillations would be limited in very large reservoirs as the viscosity of the fluid should help dampen the rise in the pressure due to the oscillating tubes of finite thickness.
3. For annular experiments, larger outer tubes could be used in order to minimize the effect of pressure induced oscillations. This would also aid in observing the second crossover at a lower frequency than the current setup will allow.

Some other considerations for the experiments are for each dilute species to have its own mass flow controller. This would more reliably keep the inlet concentration constant and more accurately keep the same inlet concentration from run to run. Transparent oscillation tubes could replace the aluminum tubes currently used so that tracer experiments could be done to analyze the flow in the tubes. A more effective method of positioning the tubes at the start of a phase angle experiment would also decrease some error involved during these types of experiments. Currently, the initial position is determined by eye, but a computer could set the initial position in some way.

6.3 Future Work in Oscillating Flows

Oscillating flows have a wide range of possibilities. One area of interest not in the scope of this thesis is in non-laminar flow. All theoretical predictions are based on a laminar assumption, but the effect of the flow being non-laminar on the mass transfer and separation is worth consideration. This would require a numerical calculation and would be based on the Kelvin-Helmholtz instability present in inviscid flows. Also, the Soret effect, or the effect of thermodiffusion, could be applied in order to enhance the mass transfer and/or separation of species. Most of this work has also focused on gases, but it can also be applied to liquid separation. The models hold true for either gases or liquids,

but another apparatus is required to perform experiments. As the ratio of the kinematic viscosity to the diffusion coefficient in liquids is very high, solutal convection may be a factor. Separation of micron-sized particles from the air could also be enhanced using pulsatile flows. This is of interest to the space program in their self-sustaining environments and to the semiconductor industry in their clean rooms. Finally, periodic flow could also be used in heat transfer problems [13, 28] because of their similarity to mass transfer problems. The findings of this thesis could perhaps aid in the optimization of various applications involving the removal of heat.

Another class of problems involving pulsatile flows would be in the area of biological fluid mechanics. In the body, the blood undergoes a periodic motion resulting from the pressure oscillations generated by the heart as it moves through the arteries and veins. An added twist is the fact that the walls of the arteries are elastic. This would require a new type of calculation to model the flow because of the change in the boundary conditions at the walls of elastic tubes. Some have already modeled the flow profile in these type of systems [29], but the mass transfer and separation has yet to be studied in these types of periodic biological flows.

Finally, a periodic displacement on the boundary, or wavy-walled tubes, is another problem of consideration. It has been suggested that a possible resonance could occur between the period of the boundary displacement and the period of the oscillations to give the greatest mass transfer and separation. These are only a few examples that I have considered as possible problems one could study in future work in pulsatile flows.

APPENDIX A VELOCITY AND CONCENTRATION CONSTANTS

Let

$$M = \frac{\omega h i \sqrt{\frac{80}{v}}}{\left(2 - h i \sqrt{\frac{80}{v}}\right) e^{h i \sqrt{\frac{80}{v}}} + \left(2 + h i \sqrt{\frac{80}{v}}\right) e^{-h i \sqrt{\frac{80}{v}}} - 4}$$

and

$$N = \frac{i \frac{v}{D} h \sqrt{\frac{160}{v}}}{L\left(\frac{v}{D} - 1\right) \left[\left(2 - h \sqrt{\frac{160}{v}}\right) e^{h \sqrt{\frac{160}{v}}} + \left(2 + h \sqrt{\frac{160}{v}}\right) e^{-h \sqrt{\frac{160}{v}}} - 4 \right]}$$

This makes

$$\psi_1 = M \left(1 - e^{-h \sqrt{\frac{80}{v}}} \right)$$

$$\psi_2 = M \left(e^{h \sqrt{\frac{80}{v}}} - 1 \right)$$

$$\psi_3 = M \left(e^{-h \sqrt{\frac{80}{v}}} - e^{h \sqrt{\frac{80}{v}}} \right)$$

$$\phi_1 = N \left[\frac{\left(1 - e^{-h \sqrt{\frac{80}{v}}} \right) \left(e^{h \sqrt{\frac{80}{v}}} - e^{h \sqrt{\frac{80}{D}}} \right) + \left(e^{h \sqrt{\frac{80}{v}}} - 1 \right) \left(e^{h \sqrt{\frac{80}{D}}} - e^{-h \sqrt{\frac{80}{v}}} \right)}{e^{-h \sqrt{\frac{80}{D}}} - e^{h \sqrt{\frac{80}{D}}}} \right] \left(\frac{D}{v} \right)^{\frac{1}{2}}$$

$$\phi_2 = N \left[\frac{\left(1 - e^{-h \sqrt{\frac{80}{v}}} \right) \left(e^{h \sqrt{\frac{80}{v}}} - e^{-h \sqrt{\frac{80}{D}}} \right) + \left(e^{h \sqrt{\frac{80}{v}}} - 1 \right) \left(e^{-h \sqrt{\frac{80}{D}}} - e^{-h \sqrt{\frac{80}{v}}} \right)}{e^{-h \sqrt{\frac{80}{D}}} - e^{h \sqrt{\frac{80}{D}}}} \right] \left(\frac{D}{v} \right)^{\frac{1}{2}}$$

$$\phi_3 = N \left(e^{h \sqrt{\frac{80}{v}}} - 1 \right)$$

$$\phi_4 = N \left(1 - e^{-h \sqrt{\frac{80}{v}}} \right)$$

$$\phi_5 = N \left(e^{h \sqrt{\frac{80}{v}}} - e^{-h \sqrt{\frac{80}{v}}} \right) \left(\frac{D}{v} - 1 \right)$$

APPENDIX B TOTAL MASS TRANSFER FOR BOUNDARY DRIVEN OPEN TUBES

Let

$$k = \sqrt{-\frac{I\omega}{D}}$$

Then

$$\bar{Q} = -\frac{D(c_2 - c_1)\pi R^2}{L} + \frac{\omega^2 A^2 (c_2 - c_1)\pi R}{4DL} \Re \left\{ \frac{i\lambda J_1(i\lambda R) [kJ_0(\lambda R)J_1(kR) - \lambda J_1(\lambda R)J_0(kR)]}{kJ_0(i\lambda R)J_0(\lambda R)J_1(kR)(k^4 - \lambda^4)} \right\}$$

APPENDIX C
ECCENTRICITY GEOMETRY CONSTANTS

Order ϵ^0 Velocity Constants

$$k_{01} = \frac{\frac{1}{4} A \omega [Y_0(i\lambda R_0) - Y_0(i\lambda R_{out})]}{Y_0(i\lambda R_0)J_0(i\lambda R_{out}) - Y_0(i\lambda R_{out})J_0(i\lambda R_0)}$$

$$k_{02} = \frac{\frac{1}{4} A \omega [J_0(i\lambda R_0) - J_0(i\lambda R_{out})]}{Y_0(i\lambda R_{out})J_0(i\lambda R_0) - Y_0(i\lambda R_0)J_0(i\lambda R_{out})}$$

$$k_{03} = \frac{\frac{1}{4} A \omega [Y_0(\lambda R_0) - Y_0(\lambda R_{out})]}{Y_0(\lambda R_0)J_0(\lambda R_{out}) - Y_0(\lambda R_{out})J_0(\lambda R_0)}$$

$$k_{04} = \frac{\frac{1}{4} A \omega [J_0(\lambda R_0) - J_0(\lambda R_{out})]}{Y_0(\lambda R_{out})J_0(\lambda R_0) - Y_0(\lambda R_0)J_0(\lambda R_{out})}$$

Order ϵ^1 Velocity Constants

$$k_{11} = \frac{i\lambda Y_1(i\lambda R_{out})[k_{01}J_1(i\lambda R_0) + k_{02}Y_1(i\lambda R_0)]}{Y_1(i\lambda R_{out})J_1(i\lambda R_0) - J_1(i\lambda R_{out})Y_1(i\lambda R_0)}$$

$$k_{12} = \frac{i\lambda J_1(i\lambda R_{out})[k_{01}J_1(i\lambda R_0) + k_{02}Y_1(i\lambda R_0)]}{J_1(i\lambda R_{out})Y_1(i\lambda R_0) - Y_1(i\lambda R_{out})J_1(i\lambda R_0)}$$

$$k_{13} = \frac{\lambda Y_1(\lambda R_{out})[k_{01}J_1(\lambda R_0) + k_{02}Y_1(\lambda R_0)]}{Y_1(\lambda R_{out})J_1(\lambda R_0) - J_1(\lambda R_{out})Y_1(\lambda R_0)}$$

$$k_{14} = \frac{\lambda J_1(\lambda R_{out})[k_{01}J_1(\lambda R_0) + k_{02}Y_1(\lambda R_0)]}{J_1(\lambda R_{out})Y_1(\lambda R_0) - Y_1(\lambda R_{out})J_1(\lambda R_0)}$$

Order ε^2 Velocity Constants

$$k_{21} = \frac{\frac{1}{2}\lambda^2 k_{01} Y_2(i\lambda R_{out}) J_0(i\lambda R_0) + \frac{1}{2}\lambda^2 k_{02} Y_2(i\lambda R_{out}) Y_0(i\lambda R_0) + k_{11} Y_2(i\lambda R_{out}) \left[i\lambda J_0(i\lambda R_0) - \frac{1}{R_0} J_1(i\lambda R_0) \right] + k_{12} Y_2(i\lambda R_{out}) \left[i\lambda Y_0(i\lambda R_0) - \frac{1}{R_0} Y_1(i\lambda R_0) \right]}{J_2(i\lambda R_{out}) Y_2(i\lambda R_0) - J_2(i\lambda R_0) Y_2(i\lambda R_{out})}$$

$$k_{22} = \frac{\frac{1}{2}\lambda^2 k_{01} J_2(i\lambda R_{out}) J_0(i\lambda R_0) + \frac{1}{2}\lambda^2 k_{02} J_2(i\lambda R_{out}) Y_0(i\lambda R_0) + k_{11} J_2(i\lambda R_{out}) \left[i\lambda J_0(i\lambda R_0) - \frac{1}{R_0} J_1(i\lambda R_0) \right] + k_{12} J_2(i\lambda R_{out}) \left[i\lambda Y_0(i\lambda R_0) - \frac{1}{R_0} Y_1(i\lambda R_0) \right]}{J_2(i\lambda R_0) Y_2(i\lambda R_{out}) - J_2(i\lambda R_{out}) Y_2(i\lambda R_0)}$$

$$k_{23} = \frac{\frac{1}{2}\lambda^2 k_{03} Y_2(\lambda R_{out}) J_0(\lambda R_0) + \frac{1}{2}\lambda^2 k_{04} Y_2(\lambda R_{out}) Y_0(\lambda R_0) + k_{13} Y_2(\lambda R_{out}) \left[\frac{1}{R_0} J_1(\lambda R_0) - \lambda J_0(i\lambda R_0) \right] + k_{14} Y_2(\lambda R_{out}) \left[\frac{1}{R_0} Y_1(\lambda R_0) - \lambda Y_0(\lambda R_0) \right]}{J_2(\lambda R_0) Y_2(\lambda R_{out}) - J_2(\lambda R_{out}) Y_2(\lambda R_0)}$$

$$k_{24} = \frac{\frac{1}{2}\lambda^2 k_{03} J_2(\lambda R_{out}) J_0(\lambda R_0) + \frac{1}{2}\lambda^2 k_{04} J_2(\lambda R_{out}) Y_0(\lambda R_0) + k_{13} J_2(\lambda R_{out}) \left[\frac{1}{R_0} J_1(\lambda R_0) - \lambda J_0(i\lambda R_0) \right] + k_{14} J_2(\lambda R_{out}) \left[\frac{1}{R_0} Y_1(\lambda R_0) - \lambda Y_0(\lambda R_0) \right]}{J_2(\lambda R_{out}) Y_2(\lambda R_0) - J_2(\lambda R_0) Y_2(\lambda R_{out})}$$

$$a_{21} = \frac{\frac{1}{2} k_{01} Y_0(i\lambda R_{out}) \left[\lambda^2 J_0(i\lambda R_0) + \frac{2i\lambda}{R_0} J_1(i\lambda R_0) \right] + \frac{1}{2} k_{02} J_0(i\lambda R_{out}) \left[\lambda^2 Y_0(i\lambda R_0) + \frac{2i\lambda}{R_0} Y_1(i\lambda R_0) \right] + k_{11} J_0(i\lambda R_{out}) \left[i\lambda J_0(i\lambda R_0) - \frac{1}{R_0} J_1(i\lambda R_0) \right] + k_{12} J_0(i\lambda R_{out}) \left[i\lambda Y_0(i\lambda R_0) - \frac{1}{R_0} Y_1(i\lambda R_0) \right]}{J_0(i\lambda R_{out}) Y_0(i\lambda R_0) - J_0(i\lambda R_0) Y_0(i\lambda R_{out})}$$

$$a_{22} = \frac{\frac{1}{2} k_{01} J_0(i\lambda R_{out}) \left[\lambda^2 J_0(i\lambda R_0) + \frac{2i\lambda}{R_0} J_1(i\lambda R_0) \right] + \frac{1}{2} k_{02} J_0(i\lambda R_{out}) \left[\lambda^2 Y_0(i\lambda R_0) + \frac{2i\lambda}{R_0} Y_1(i\lambda R_0) \right] + k_{11} J_0(i\lambda R_{out}) \left[i\lambda J_0(i\lambda R_0) - \frac{1}{R_0} J_1(i\lambda R_0) \right] + k_{12} J_0(i\lambda R_{out}) \left[i\lambda Y_0(i\lambda R_0) - \frac{1}{R_0} Y_1(i\lambda R_0) \right]}{J_0(i\lambda R_0) Y_0(i\lambda R_{out}) - J_0(i\lambda R_{out}) Y_0(i\lambda R_0)}$$

$$a_{23} = \frac{\frac{1}{2} k_{03} Y_0(\lambda R_{out}) \left[\lambda^2 J_0(\lambda R_0) - \frac{2\lambda}{R_0} J_1(\lambda R_0) \right] + \frac{1}{2} k_{04} Y_0(\lambda R_{out}) \left[\lambda^2 Y_0(\lambda R_0) - \frac{2\lambda}{R_0} Y_1(\lambda R_0) \right] + k_{13} Y_0(\lambda R_{out}) \left[\frac{1}{R_0} J_1(\lambda R_0) - \lambda J_0(\lambda R_0) \right] + k_{14} Y_0(\lambda R_{out}) \left[\frac{1}{R_0} Y_1(\lambda R_0) - \lambda Y_0(\lambda R_0) \right]}{J_0(\lambda R_0) Y_0(\lambda R_{out}) - J_0(\lambda R_{out}) Y_0(\lambda R_0)}$$

$$a_{24} = \frac{\frac{1}{2} k_{03} J_0(\lambda R_{out}) \left[\lambda^2 J_0(\lambda R_0) - \frac{2\lambda}{R_0} J_1(\lambda R_0) \right] + \frac{1}{2} k_{04} J_0(\lambda R_{out}) \left[\lambda^2 Y_0(\lambda R_0) - \frac{2\lambda}{R_0} Y_1(\lambda R_0) \right] + k_{13} J_0(\lambda R_{out}) \left[\frac{1}{R_0} J_1(\lambda R_0) - \lambda J_0(\lambda R_0) \right] + k_{14} J_0(\lambda R_{out}) \left[\frac{1}{R_0} Y_1(\lambda R_0) - \lambda Y_0(\lambda R_0) \right]}{J_0(\lambda R_{out}) Y_0(\lambda R_0) - J_0(\lambda R_0) Y_0(\lambda R_{out})}$$

Order ϵ^0 Concentration Constants

$$m_{01} = \frac{\frac{\Delta C}{DL} \frac{\lambda}{\lambda^2 - \alpha^2} [k_{01} J_1(i\lambda R_0) + k_{02} Y_1(i\lambda R_0)]}{\alpha [J_1(i\alpha R_{out}) Y_1(i\alpha R_0) - Y_1(i\alpha R_{out}) J_1(i\alpha R_0)]} Y_1(i\alpha R_{out}) - \frac{\frac{\Delta C}{DL} \frac{\lambda}{\lambda^2 - \alpha^2} [k_{01} J_1(i\lambda R_{out}) + k_{02} Y_1(i\lambda R_{out})]}{\alpha [J_1(i\alpha R_{out}) Y_1(i\alpha R_0) - Y_1(i\alpha R_{out}) J_1(i\alpha R_0)]} Y_1(i\alpha R_0)$$

$$m_{02} = \frac{\frac{\Delta C}{DL} \frac{\lambda}{\lambda^2 - \alpha^2} [k_{01} J_1(i\lambda R_{out}) + k_{02} Y_1(i\lambda R_{out})]}{\alpha [J_1(i\alpha R_{out}) Y_1(i\alpha R_0) - Y_1(i\alpha R_{out}) J_1(i\alpha R_0)]} J_1(i\alpha R_0) - \frac{\frac{\Delta C}{DL} \frac{\lambda}{\lambda^2 - \alpha^2} [k_{01} J_1(i\lambda R_0) + k_{02} Y_1(i\lambda R_0)]}{\alpha [J_1(i\alpha R_{out}) Y_1(i\alpha R_0) - Y_1(i\alpha R_{out}) J_1(i\alpha R_0)]} J_1(i\alpha R_{out})$$

$$m_{03} = \frac{\frac{\Delta C}{DL} \frac{\lambda}{\alpha^2 - \lambda^2} [k_{03} J_1(\lambda R_0) + k_{04} Y_1(\lambda R_0)]}{\alpha [J_1(\alpha R_{out}) Y_1(\alpha R_0) - Y_1(\alpha R_{out}) J_1(\alpha R_0)]} Y_1(\alpha R_{out}) - \frac{\frac{\Delta C}{DL} \frac{\lambda}{\alpha^2 - \lambda^2} [k_{03} J_1(\lambda R_{out}) + k_{04} Y_1(\lambda R_{out})]}{\alpha [J_1(\alpha R_{out}) Y_1(\alpha R_0) - Y_1(\alpha R_{out}) J_1(\alpha R_0)]} Y_1(\alpha R_0)$$

$$m_{04} = \frac{\frac{\Delta C}{DL} \frac{\lambda}{\alpha^2 - \lambda^2} [k_{03} J_1(\lambda R_{out}) + k_{04} Y_1(\lambda R_{out})]}{\alpha [J_1(\alpha R_{out}) Y_1(\alpha R_0) - Y_1(\alpha R_{out}) J_1(\alpha R_0)]} J_1(\alpha R_0) \\ - \frac{\frac{\Delta C}{DL} \frac{\lambda}{\alpha^2 - \lambda^2} [k_{03} J_1(\lambda R_0) + k_{04} Y_1(\lambda R_0)]}{\alpha [J_1(\alpha R_{out}) Y_1(\alpha R_0) - Y_1(\alpha R_{out}) J_1(\alpha R_0)]} J_1(\alpha R_{out})$$

Order ε^1 Concentration Constants

$$\text{Let } \chi_1(\alpha R_i) = \alpha J_0(\alpha R_i) - \frac{1}{R_i} J_1(\alpha R_i); \quad \chi_2(\alpha R_i) = \alpha Y_0(\alpha R_i) - \frac{1}{R_i} Y_1(\alpha R_i)$$

$$\text{and } \chi_1(\lambda R_i) = \lambda J_0(\lambda R_i) - \frac{1}{R_i} J_1(\lambda R_i); \quad \chi_2(\lambda R_i) = \lambda Y_0(\lambda R_i) - \frac{1}{R_i} Y_1(\lambda R_i)$$

where R_i is R_0 or R_{out}

$$m_{14} = \frac{\frac{\Delta C}{DL} \frac{1}{\alpha^2 - \lambda^2} [k_{13} \chi_1(\lambda R_{out}) + k_{14} \chi_2(\alpha R_{out})]}{-\chi_1(\alpha R_{out})} \chi_1(\alpha R_0) - m_{03} \alpha \chi_1(\alpha R_0) - m_{04} \alpha \chi_2(\alpha R_0) \\ + \frac{\frac{\Delta C}{DL} \frac{1}{\alpha^2 - \lambda^2} [k_{13} \chi_1(\lambda R_0) + k_{14} \chi_2(\lambda R_0) - k_{03} \lambda \chi_1(\lambda R_0) - k_{04} \lambda \chi_2(\lambda R_0)]}{\frac{\chi_2(\alpha R_{out})}{\chi_1(\alpha R_{out})} \chi_1(\alpha R_0) - \chi_2(\alpha R_0)} \\ m_{13} = \frac{m_{14} \chi_2(\alpha R_{out})}{-\chi_1(\alpha R_{out})} + \frac{\frac{\Delta C}{DL} \frac{1}{\alpha^2 - \lambda^2} [k_{13} \chi_1(\lambda R_{out}) + k_{14} \chi_2(\alpha R_{out})]}{-\chi_1(\alpha R_{out})}$$

Order ε^2 Concentration Constants

$$\text{Let } \eta_1(\alpha R_i) = \alpha J_1(\alpha R_i) - \frac{1}{R_i} J_2(\alpha R_i); \quad \eta_2(\alpha R_i) = \alpha Y_1(\alpha R_i) - \frac{1}{R_i} Y_2(\alpha R_i)$$

$$\text{and } \eta_1(\lambda R_i) = \lambda J_1(\lambda R_i) - \frac{1}{R_i} J_2(\lambda R_i); \quad \eta_2(\lambda R_i) = \lambda Y_1(\lambda R_i) - \frac{1}{R_i} Y_2(\lambda R_i)$$

where R_i is R_0 or R_{out}

$$\begin{aligned}
& \frac{\Delta C}{DL} \frac{\lambda}{\alpha^2 - \lambda^2} [a_{23} J_1(\lambda R_{out}) + a_{24} Y_1(\lambda R_{out})] \frac{J_1(\alpha R_0)}{J_1(\alpha R_{out})} - \frac{\Delta C}{DL} \frac{\lambda}{\alpha^2 - \lambda^2} [a_{23} J_1(\lambda R_0) + a_{24} Y_1(\lambda R_0)] \\
& + \frac{1}{2} m_{03} \alpha^2 \eta_1(\alpha R_0) + \frac{1}{2} m_{04} \alpha^2 \eta_2(\alpha R_0) + \frac{\Delta C}{2DL} \frac{\lambda^2}{\alpha^2 - \lambda^2} [k_{03} \eta_1(\lambda R_0) + k_{04} \eta_2(\lambda R_0)] \\
& - m_{13} \alpha \eta_1(\alpha R_0) - m_{14} \alpha \eta_2(\alpha R_0) - \frac{\Delta C}{DL} \frac{\lambda}{\alpha^2 - \lambda^2} [k_{13} \eta_1(\lambda R_0) + k_{14} \eta_2(\lambda R_0)] \\
& + \frac{m_{03} \alpha}{2R_0} \chi_1(\alpha R_0) + \frac{m_{04} \alpha}{2R_0} \chi_2(\alpha R_0) + \frac{\Delta C}{2DL R_0} \frac{\lambda}{\alpha^2 - \lambda^2} [k_{03} \chi_1(\lambda R_0) + k_{04} \chi_2(\lambda R_0)] \\
b_{24} = & \frac{\alpha Y_1(\alpha R_0) - \frac{\alpha Y_1(\alpha R_{out})}{J_1(\alpha R_{out})} J_1(\alpha R_0)}{\alpha Y_1(\alpha R_0) - \frac{\alpha Y_1(\alpha R_{out})}{J_1(\alpha R_{out})} J_1(\alpha R_0)} \\
b_{23} = & \frac{-b_{24} Y_1(\alpha R_{out})}{J_1(\alpha R_{out})} - \frac{\Delta C}{DL} \frac{a_{23}}{\alpha^2 - \lambda^2} \frac{\lambda}{\alpha} \frac{J_1(\lambda R_{out})}{J_1(\alpha R_{out})} - \frac{\Delta C}{DL} \frac{a_{24}}{\alpha^2 - \lambda^2} \frac{\lambda}{\alpha} \frac{Y_1(\lambda R_{out})}{J_1(\alpha R_{out})}
\end{aligned}$$

APPENDIX D DISTRIBUTION OF AREA IN AN ECCENTRIC ANNULUS

In an annular geometry, when the inner cylinder is shifted from the center of the outer cylinder, the region that has become “thicker” than the area of the centered annulus is greater than the region that is now “thinner”. To find the area of the eccentric annulus, one must calculate

$$\text{Area} = \pi(R_{out}^2 - R_{in}^2) \quad (\text{D.1})$$

To prove that there is a greater “thicker” region, the new inner radius for the eccentric annulus must be less along the circumference of the inner cylinder than the inner radius for the centered annulus. If $R_{in,0} = R_{in}$, then from Equation 3.10 this would occur at an angle

$$\theta = \cos^{-1}\left(\frac{\varepsilon}{2R_{in,0}}\right) \quad (\text{D.2})$$

As ε is smaller than $R_{in,0}$, the value for θ where the radii will be the same will occur near $\pi/2$ but between $0 < \theta < \frac{\pi}{2}$. This shows that there is a greater region in which $R_{out} - R_{in}$ is greater than $R_{out} - R_{in,0}$, and thus the thicker region is greater.

APPENDIX E GAS FLOW BETWEEN OSCILLATING FLAT PLATES

This appendix is a report done by Brad Ciccirelli during his time as an undergraduate in chemical engineering. This calculation was done as a part of his work in our laboratory.

Consider two parallel flat plates as shown below in Figure E.1. Suppose that the two plates move solely in the x -direction with a velocity given by $V_x = \frac{1}{2} A \omega \cos(\omega t)$. The motion of a gaseous species present between the plates can be described by the Navier-Stokes equation, which in this case reduces to

$$\rho \frac{\partial V_x}{\partial t} = \mu \frac{\partial^2 V_x}{\partial y^2} \quad (\text{E.1})$$

The solution to this equation will be of the form

$$V_x(y, t) = f(y) \cos(\omega t) = \hat{v}_x(y) e^{i\omega t} + \tilde{v}_x(y) e^{-i\omega t} \quad (\text{E.2})$$

where $\hat{v}_x(y)$ and $\tilde{v}_x(y)$ are complex conjugates. Application of the no-slip boundary

conditions at the plate walls and introduction of the kinematic viscosity $\nu = \frac{\mu}{\rho}$ yields

the solution

$$\hat{v}_x(y) = \frac{A \omega \left(e^{(i+1)\sqrt{\frac{\omega}{2\nu}}y} + e^{-(i+1)\sqrt{\frac{\omega}{2\nu}}y} \right)}{4 \left(e^{(i+1)\sqrt{\frac{\omega}{2\nu}}h} + e^{-(i+1)\sqrt{\frac{\omega}{2\nu}}h} \right)} \quad (\text{E.3})$$

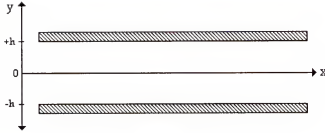


Figure E.1. Two parallel flat plates.

Solving for $\tilde{v}_x(y)$ in a similar manner and combining the solutions, we find

$$V_x(y, t) = \frac{\frac{1}{4} A \omega e^{i \omega t} \left(e^{(i+1) \sqrt{\frac{\omega}{2\nu}} y} + e^{-(i+1) \sqrt{\frac{\omega}{2\nu}} y} \right)}{\left(e^{(i+1) \sqrt{\frac{\omega}{2\nu}} h} + e^{-(i+1) \sqrt{\frac{\omega}{2\nu}} h} \right)} + \frac{\frac{1}{4} A \omega e^{-i \omega t} \left(e^{(i-1) \sqrt{\frac{\omega}{2\nu}} y} + e^{(1-i) \sqrt{\frac{\omega}{2\nu}} y} \right)}{\left(e^{(i-1) \sqrt{\frac{\omega}{2\nu}} h} + e^{(1-i) \sqrt{\frac{\omega}{2\nu}} h} \right)} \quad (\text{E.4})$$

Now that the velocity profile is known, the concentration profile can be found

from a species balance

$$\frac{\partial c}{\partial t} + V_x \frac{\partial c}{\partial x} = D \frac{\partial^2 c}{\partial x^2} + D \frac{\partial^2 c}{\partial y^2} \quad (\text{E.5})$$

where c is the concentration of the gaseous species and D is its molecular diffusivity. If

we assume a constant concentration gradient in the x -direction

$$\frac{\partial c}{\partial x} = \frac{\Delta c}{L} = \gamma \quad (\text{E.6})$$

we can express the concentration profile as

$$c(x, y, t) = \gamma x + \hat{c}(y) e^{i \omega t} + \tilde{c}(y) e^{-i \omega t} \quad (\text{E.7})$$

where $\hat{c}(y)$ and $\tilde{c}(y)$ are complex conjugates. Application of the impermeable wall boundary conditions

$$\left(\frac{\partial c}{\partial y}\right)_{y=h} = \left(\frac{\partial c}{\partial y}\right)_{y=-h} = 0 \quad (\text{E.8})$$

gives the solution

$$\hat{c}(y) = \frac{\Psi \alpha (e^{\alpha h} - e^{-\alpha h}) \left(e^{\sqrt{\frac{i\omega}{D}} y} + e^{-\sqrt{\frac{i\omega}{D}} y} \right)}{\sqrt{\frac{i\omega}{D}} (D\alpha^2 - i\omega) \left(e^{-\sqrt{\frac{i\omega}{D}} h} - e^{\sqrt{\frac{i\omega}{D}} h} \right)} + \frac{\Psi}{(D\alpha^2 - i\omega)} (e^{\alpha y} + e^{-\alpha y}) \quad (\text{E.9})$$

where

$$\Psi = \frac{A\omega\gamma}{4(e^{\alpha h} + e^{-\alpha h})} \quad (\text{E.10})$$

and

$$\alpha = \sqrt{\frac{i\omega}{\nu}} \quad (\text{E.11})$$

The solution for $\hat{c}(y)$ is found in a similar manner.

Now consider non-oscillating plates with pressure driven flow between them. The Navier-Stokes equation for this case reduces to

$$0 = -\frac{\partial p}{\partial x} + \mu \frac{\partial^2 V_x}{\partial y^2} \quad (\text{E.12})$$

Treating $\frac{\partial p}{\partial x}$ as a constant and applying the no-slip boundary conditions at the plate walls

$$(V_x)_{y=h} = (V_x)_{y=-h} = 0 \quad (\text{E.13})$$

gives

$$V_x = \frac{1}{2\mu} \left(\frac{\partial p}{\partial x} \right) (y^2 - h^2) \quad (\text{E.14})$$

The mean velocity $\bar{V}$ for this parabolic profile is given by

$$\bar{V} = \frac{2}{3} V_{\max} = \frac{-h^2}{3\mu} \left(\frac{\partial p}{\partial x} \right) \quad (\text{E.15})$$

A velocity V' relative to the mean velocity can be defined as $V' = V_x - \bar{V}$. This is the velocity seen by an observer moving with the mean flow. Expressed mathematically, this is

$$V' = V_x - \bar{V} = \frac{1}{2\mu} \left(\frac{\partial p}{\partial x} \right) (y^2 - h^2) + \frac{h^2}{3\mu} \left(\frac{\partial p}{\partial x} \right) = \frac{1}{6\mu} \left(\frac{\partial p}{\partial x} \right) (3y^2 - h^2) \quad (\text{E.16})$$

Using this relative velocity V' , a species balance can be written. This expression for V' corresponds to steady-state flow and therefore yields a concentration profile that is not time dependent, i.e. $\frac{\partial c}{\partial t} = 0$. Also, if diffusion in the x -direction is considered small

compared to diffusion in the y -direction, i.e. $D \frac{\partial^2 c}{\partial x^2} \ll D \frac{\partial^2 c}{\partial y^2}$, then the species balance

may be written as

$$V' \frac{\partial c}{\partial x} = D \frac{\partial^2 c}{\partial y^2} \quad (\text{E.17})$$

Using the impermeable wall boundary conditions mentioned earlier and solving,

$$c = \Delta c + \frac{\gamma}{24\mu D} \left(\frac{\partial p}{\partial x} \right) y^4 - \frac{\gamma h^2}{12\mu D} \left(\frac{\partial p}{\partial x} \right) y^2 \quad (\text{E.18})$$

Now consider a scenario in which there is gas flow due to the oscillating plates and counterflow against the concentration gradient due to a pressure drop along the length of the plates. This is a combination of the two previously considered cases. Due

to the assumptions that the concentration and pressure gradients are constant, the velocity and concentration profiles are additive.

$$V_x = V^I + V^{II} \quad \text{and} \quad C = C^I + C^{II} \quad (\text{E.19,E.20})$$

The superscripts I and II correspond to the purely oscillatory case and the purely pressure-driven case respectively. The molar flux J_x of a species is given by

$$J_x = V_x C - D \frac{\partial C}{\partial x} \quad (\text{E.21})$$

For the combined scenario,

$$\begin{aligned} J_x &= (V^I + V^{II})(C^I + C^{II}) - D\gamma \\ &= (\hat{v}_x e^{i\omega t} + \bar{v}_x e^{-i\omega t} + V')(\gamma x + \hat{c} e^{i\omega t} + \bar{c} e^{-i\omega t} + C^{II}) - D\gamma \end{aligned} \quad (\text{E.22})$$

The time averaged molar flux $\bar{J}_x$ is defined as

$$\bar{J}_x = \frac{1}{P} \int_0^P J_x dt \quad (\text{E.23})$$

where

$$P = \text{period} = \frac{2\pi}{\omega} \quad (\text{E.24})$$

For the scenario now considered,

$$\bar{J}_x = \bar{c} \hat{v}_x + \hat{c} \bar{v}_x + \gamma x V' + C^{II} V' - D\gamma = 2\text{Re}\{\hat{c} \bar{v}_x\} + \gamma x V' + C^{II} V' - D\gamma \quad (\text{E.25})$$

The time- and spatially-averaged mass transfer $\bar{Q}_x$ is given by

$$\bar{Q}_x = \frac{1}{A} \iint_A \bar{J}_x dA \quad (\text{E.26})$$

which for this case simplifies to

$$\bar{Q}_x = \frac{1}{2h} \int_{-h}^h J_x dy = \frac{1}{2h} \int_{-h}^h [2\text{Re}\{\hat{c} \hat{v}_x\} + \gamma x V' + C'' V' - D\gamma] dy \quad (\text{E.27})$$

substituting and integrating, we find that

$$\bar{Q}_x = \frac{1}{h} \text{Re} \left\{ \int_{-h}^h \hat{c} \hat{v} dy \right\} + \frac{2\gamma}{945\mu^2 D} \left(\frac{\partial p}{\partial x} \right)^2 h^6 - D\gamma \quad (\text{E.28})$$

This result leads to some interesting phenomena.

Figure E.2 is a plot of $\bar{Q}_x$ vs ω for two different values of D with all other variables held fixed, and the values of the variables are given on the next page. The plot shows multiple points of intersection between the two curves. This demonstrates that there are frequencies at which a species with a lower diffusivity actually has greater mass transfer in the x -direction.

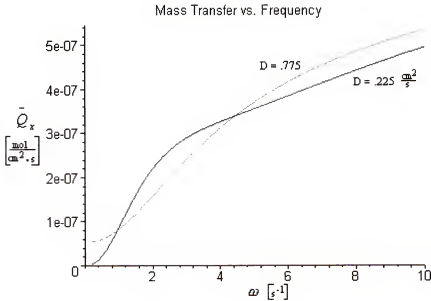


Figure E.2. Plot of $\bar{Q}_x$ vs ω with a height of 1 cm.

A plot of $\bar{Q}_x$ vs h with all other variables held fixed is shown in Figure E.3. This curve shows a maximum. This demonstrates that for a given frequency, there is an optimum plate separation for mass transfer.

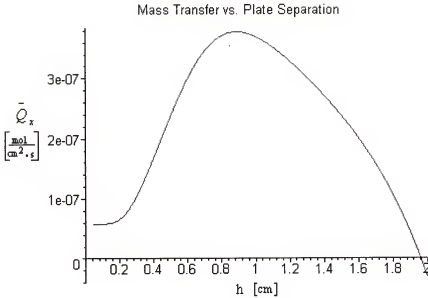


Figure E.3. Plot of $\bar{Q}_x$ vs h for a frequency of 5 rad/sec.

The laboratory setup has concentric cylinders moving in phase in the axial direction. The flow for this case is in the annular region between the two cylinders.

Typical operating conditions for his experiments are as outlined below:

A = amplitude of oscillation = 15 cm

L = length of cylinders = 134 cm

h = $\frac{1}{2}$ the difference in the cylinder radii = .25 cm

ν = kinematic viscosity of the gaseous species = .15 cm²/sec

D = diffusivity of the gaseous species = .775 cm²/sec

μ = dynamic viscosity of the gaseous species = .01813 g/cm*sec

$$\gamma = \frac{\Delta c}{L} = \text{concentration gradient} = -7.5 \times 10^{-8} \frac{\text{mol}}{\text{cm}^4}$$

$$\omega = \text{frequency of oscillation [s}^{-1}\text{]}$$

If the annular region is small compared to the outer radius, this may be approximated by flow in a channel and can be modeled with the equations derived earlier. One interesting problem of great practical importance is determining the pressure drop for a given frequency of oscillation that yields no net mass transfer. This occurs when the mass transfer due to the oscillations and pure diffusion is exactly canceled by the counterflow.

This can be found by examining a plot of $\bar{Q}_x$ vs $\frac{\partial p}{\partial x}$ for a given ω and h , as shown in Figure E.4. The x -intercept of the curve gives the critical pressure gradient at which the pressure-driven counterflow begins to overtake the boundary-driven oscillatory flow. This critical pressure gradient was calculated for various frequencies and two different values of plate separation. The results can be found in Table E.1.

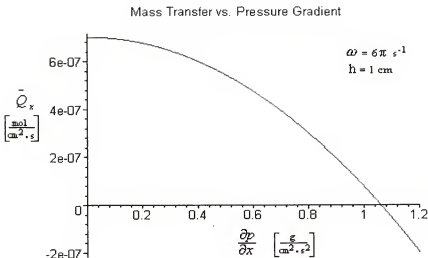


Figure E.4. Plot of $\bar{Q}_x$ vs $\frac{\partial p}{\partial x}$

Table E.1. Critical pressure drops for given frequencies and plate separations.

		$h = 1 \text{ cm}$			$h = .25 \text{ cm}$		
ω	RPM	$\frac{\partial p}{\partial x}$	$\frac{\partial p}{\partial x}$	Δp	$\frac{\partial p}{\partial x}$	$\frac{\partial p}{\partial x}$	Δp
$[s^{-1}]$		$\left[\frac{g}{cm^2 \cdot s^2} \right]$	$\left[\frac{atm}{cm} \right]$ $\times 10^6$	$[atm]$ $\times 10^4$	$\left[\frac{g}{cm^2 \cdot s^2} \right]$	$\left[\frac{atm}{cm} \right]$ $\times 10^4$	$[atm]$
2π	60	.82	.809	1.08	26	.257	.003
4π	120	.97	.957	1.28	46	.454	.006
6π	180	1.06	1.05	1.40	67	.661	.009
8π	240	1.14	1.13	1.51	88	.868	.012
10π	300	1.20	1.18	1.59	105	1.03	.014

For example, for a plate separation of 2 cm ($h = 1 \text{ cm}$) and oscillations of 120 RPM, a pressure drop greater than $1.28 \times 10^{-4} \text{ atm}$ will cause mass transfer due to pressure-driven flow to dominate.

This critical pressure gradient is an important parameter needed to assess experimental integrity. If this value is less than the uncertainty in the pressure measurements, then great depression or enhancement of the mass transfer can occur without detection. The results in Table E.1 show that as the plate separation is reduced, the value of the critical pressure gradient increases, allowing more tolerance for a reasonable pressure drop in experiments.

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BIOGRAPHICAL SKETCH

The author was born on the army base in Fort Hood, TX. He obtained his undergraduate degree in chemical engineering at Stanford University in 1996. In the same year, he began his graduate studies at the University of Florida. In 1997 he married Susan Elizabeth Brown, and they currently have two children, Yesenia Adelaida and Dakota Aaron.

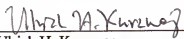
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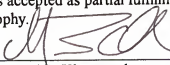

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